

Appendix A

Recipes for Surrogates, Grouts, and Other Material

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Appendix A

Recipes for Surrogates, Grouts, and Other Material

A-1. RECIPE FOR SIMULATED GROUNDWATER USED IN LEACH TESTING

Table A-1. Simulated groundwater composition.

Water Characteristics	Value (mg/kg) ^a
Sulfate	88
Chloride	124
Nitrate	5
Sodium	26
Bicarbonate	71
Potassium	4
Calcium	70
Magnesium	22
Total dissolved solids	410
Conductivity	567 (μs/cm)
Acidity	8 (pH units)

a. Unless otherwise indicated.

The following is the Idaho National Laboratory synthetic groundwater recipe (based on chemical analysis of groundwater samples collected at the Idaho National Laboratory Site).

Table A-2. Recipe for simulated groundwater based on a volume of 50 liters of nanopure water.

Compound	Formula	Mass (g)
Potassium nitrate	KNO ₃	0.20
Magnesium sulfate	MgSO ₄	5.50
Calcium chloride	CaCl ₂	9.70
Sodium nitrate	NaNO ₃	0.17
Sodium bicarbonate	NaHCO ₃	4.62
Potassium bicarbonate	KHCO ₃	0.31

A-2. RECIPES (EXCLUDING RADIONUCLIDES)^a FOR NITRATE SALT SLUDGE, INORGANIC SLUDGE, AND ORGANIC SLUDGE SURROGATES

Table A-3. Subsurface Disposal Area nitrate salt sludge (Rocky Flats Plant Series 745) surrogate.

Material	Historical Content (wt%)		Bench Test Batch Using Typical Salts (wt%)	
	1978	1992		
Sodium	16.20	23.90	Sodium nitrite	0.50
Potassium	14.50	8.60	Potassium nitrate	30.00
Nitrate	62.30	54.90	Sodium nitrate	60.00
Chlorine	2.80	3.10	Sodium chloride	3.00
Fluorine	0.30	0.60	Sodium fluoride	0.50
Phosphate	1.30	1.40	Sodium phosphate	1.00
Sulfate	3.60	3.60	Sodium sulfate	3.00
Carbonate	N/A	0.40	Sodium bicarbonate	1.00
Chromium	0.03	0.04	Chromium nitrate	0.04
Organics	N/A	1.00	EDTA	1.00
Water	N/A	2.00	Water	2.00
Total	99.10	99.50	Percent water added to above mix	100

EDTA = ethylene diaminetetraacetic

Table A-4. Subsurface Disposal Area organic sludge (Rocky Flats Plant Series 743) surrogate.

Material	Specific Gravity ^a	Historic Estimated Concentration				Selected Concentration (wt%)
		(vol%)		(wt%)		
		1981 ^b	1998 ^c	1981	1998 ^c	
Texaco Regal oil	0.87	22.0	16	20	14	29.0
Miscellaneous oil ^d	0.90	11.0 ^d	8	13	10	
Carbon tetrachloride	1.59	17.0	19	27	30	27.0
TCE ^d	1.46	4.0	5	6	6	7.0
TCA	1.44	5.0	6	9	8	9.0
PCE ^d	1.59	4.0	5	6	6	7.0
Silicon oxide	2.50	8.4	12	21	29	13.5
Oil Dri	2.30					7.3

a. The organic chlorinated solvents are over 60% denser than the oils. (Specific gravity is 1.5 versus 0.87.)

b. The year of records and estimation.

c. *Estimate of Carbon Tetrachloride in 743 Series Sludges Buried in the Subsurface Disposal Area at the Radioactive Waste Management Complex* (Miller and Navratil 1998).

d. 43% miscellaneous oil (mineral oil used for the surrogate) and solvents are listed in shipping records, of which 20% has been divided amongst TCE and PCE. The TCE and PCE are put here from the miscellaneous oil category.

PCE = tetrachloroethene

TCA = trichloroethane

TCE = trichloroethene

a. The radionuclide content was tailored to a specific task and, therefore, is covered in the materials section of the specific task.

Table A-5. Composition of Rocky Flats Plant Series 741 and 742 inorganic sludge surrogate.

Compound	Concentration (wt%)
Soil	20 ^a
Calcium carbonate	10
Water	20 ^a
Portland cement	10
Rare-earth tracer	2
Calcium nitrate	3
Sodium nitrate	10
Potassium nitrate	5
Sodium hydrogen phosphate water	20

a. Soil from the Idaho National Engineering and Environmental Laboratory Site contains 15 wt% water.

A-3. COMPOSITION AND BASIC INFORMATION ON GROUTS USED IN TESTS

Table A-6. Composition of grouts for in situ grouting.

Material	GMENT-12	U.S. Grout	TECT HG	WAXFIX
Classification	Inorganic	Inorganic	Inorganic	Organic thermo plastic
Designer	Savannah River Plant	Hess Pumice E. H. Ahrens	Ernie Carter Technologies	Ernie Carter Technologies
Designed use	Tank closure grout	Sealing fine cracks at the Waste Isolation Pilot Plant	Enhanced jet grouting fixation of mercury in soil	Stabilizing buried waste in silty clay
Base ingredient binder	56.7% ^a Portland Type V	Microfine Portland Type H	Proprietary Portland Type H	Paraffin
Pozzolanic material	8.8% GBFS ^b 3.8% silica fume ^c	Natural Idaho Pumice	Proprietary pulverized hematite filler	Proprietary fillers, sodium borate
Metal fixant	0.1% sodium thiosulfide	0	Sodium sulfide ^d	Sodium sulfide ^e
Plasticizer ^f	0.46%	1.8% Distil ^g	Proprietary	N/A
Set retarder ^h	0.15%	0	Proprietary	N/A
Water ⁱ	30.2%	37%	Proprietary	0 ^j

a. All percentages are weight percent.

b. Ground blast furnace slag, ASTM-989 grade or better.

c. ASTM C1240 slurry form.

d. Main part of the proprietary mercury fixation formulation.

e. Possible if metal precipitation is desired.

f. Plasticizer or high-range water reducer ASTM C494 Type F.

g. Powdered water reducer.

h. ASTM C494 Type C or D.

i. Water amount listed refers to the recommended total weight percent of water content in the grout. The amount of water added to the grout can be adjusted to accommodate the expected water content of the waste.

j. No water is required for WAXFIX. WAXFIX may drive some water from the waste to the extent that the raised temperature volatilizes it. Some water may remain suspended in the wax as it cools. This water does not affect the WAXFIX properties any more than the properties of any of the grouts used.

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Table A-7. Composition of solidification agents for Pad A salt solidification.

Material	Grout		
	Saltstone	Polysiloxane	WAXFIX
Classification	Inorganic	Organic thermo set	Organic thermo plastic
Designer	Savannah River Plant	Technology Visions Group	Ernie Carter Technologies
Designed use	Liquid salt waste	Encapsulate large quantities of salts	Stabilizing buried waste in silty clay
Base ingredient binder	3.3 % ^a Portland Type II	Dimethyl-Polysiloxane	Paraffin
Pozzolanic material	27.7 % slag ^b , 27.7 % fly ash ^c	Proprietary fillers	Proprietary fillers, sodium borate
Metal fixant	Reductants	0	Sodium sulfide
Plasticizer ^d	Recommend ^e	N/A	N/A
Set retarder	Recommend ^e	N/A	N/A
Water	41.3% ^e	0	0

a. All percentages are weight percent

b. Grade 120 slag

c. Class F fly ash

d. Plasticizer or high-range water reducer ASTM C494 Type F

e. Water amount listed that is present in waste and may be reduced with plasticizer or combination set retarder.

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Table A-8. Grout formulations for nontransuranic waste.^a

Waste Form	Nontransuranic Waste (wt%)	Water (wt%)	Cement (with plasticizer) (wt%)	Slag (wt%)	Fly Ash (wt%)	Thiosulfate (wt%)	Mix Density (g/cm ³)
Cement	46.8	8.7	44.5	0	0	0	2.09
Cement, slag	53.9	7.5	19.4	19.2	0	0	2.07
Cement, fly ash	43.4	10	23.2	0	23.2	0	2.08
Cement, fly ash, thiosulfate	42.3	9.7	24.4	0	23.4	0.1	2.006
Cement, slag, sodium thiosulfate	46.6	10	22	21.7	0	0.1	2.007
Blank (cement, fly ash, slag, sodium thiosulfate)	Cold soil 44	24	10.6	10.4	10.4	0.05	2.005

a. The recipes in this table were used in Appendixes P, R, S, and T.

A-4. REFERENCES

Miller, Eric C. and James D. Navratil, 1998, *Estimate of Carbon Tetrachloride in 743 Series Sludges Buried in the Subsurface Disposal Area at the Radioactive Waste Management Complex*, INEEL/EXT-98-00112, Rev. 0, Idaho National Engineering and Environmental Laboratory.

Yancey, Neal A., Peter G. Shaw, David F. Nickelson, Gretchen E. Matthern, and Guy G. Loomis, 2003, *Test Plan for the Evaluation of In Situ Thermal Desorption and Grouting Technologies for Operable Unit 7-13/14*, INEEL/EXT-03-00059, Rev. 0, Idaho National Engineering and Environmental Laboratory, Idaho Completion Project.

Appendix B

Compressive-Strength Tests—Part 1

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Appendix B

Compressive-Strength Tests—Part 1

Samples of neat grout mixed with surrogates were tested for unconfined compressive strength. For full-scale in situ grouting (ISG) at the Subsurface Disposal Area (SDA), the grouted-waste monolith must provide a stable foundation for material placed on it, including an engineered cap. Collapse and subsidence of soil into subsurface voids occur at the SDA during wet conditions and have the potential to compromise the integrity of a cap. Soil subsidence affects the hydraulic properties of the SDA by causing ponding of surface water, which also can lead to an increase in the development of permeable pathways to the waste. Compressive-strength testing was used as a means of selecting grout and waste combinations that, when placed as a monolith, will support an engineered cover without subsidence. Grouting also may be used to place columns in the waste to support a cap. In this case, the compressive strength would need to be higher than for a monolith. While the required compressive strength has not yet been determined, these data provide useful information for selecting grouts for this application.

In previous grout evaluations (Loomis et al. 2003), unconfined compressive strength of 250 psi was used as performance criteria. While it is recognized that compressive strength does not necessarily correlate with contaminant immobilization, it is a semiquantitative indicator of overall grout performance. American National Standards Institute leach testing requires samples, which are stand-alone monoliths. Samples with unconfined compressive strength of greater than 250 psi meet this requirement. The 250-psi criterion was used in this study to be consistent with previous work and to support the American National Standards Institute leach tests.

The results of compressive-strength testing were used to select waste loadings for the remaining tests performed in this work. Where data were available from previous testing (Loomis et al. 2003), they were used. Where data were not available, they were generated as a part of this work.

The interference of soil, nitrate salt, and organic sludge may adversely affect grout performance. Compressive strength was tested for specially prepared grout samples mixed with these simulated interference materials at various concentrations. This comparison gives confidence in using bench-derived data to evaluate future grout types for application of ISG of buried waste. Test results and observations determine the waste-loading tolerance for grout materials and the compatibility of chosen grouts with contaminants expected in waste buried at the SDA.

B-1. TEST OBJECTIVES, RATIONALE, AND DEVIATIONS FROM TEST PLAN

The main goals for ISG are immobilization of contaminants of concern and structural stability of waste. To establish the effectiveness of ISG as a waste treatment applicable to the SDA, the resulting monolith of grout and waste must exhibit the following attributes:

- Compatibility of grout and waste. Waste matrices such as nitrate salts and organic sludges, when mixed with grout, may interfere with grout curing. Candidate grouts must be tolerant of these interferences, maintaining good compressive strength when mixed with waste.
- Structural stability of waste. The injected grout mixture must structurally stabilize buried waste to prevent site subsidence and prevent surface water ponding.

Compressive strength was used as an objective standard for defining a cohesive (that is, stand-alone) monolith and determining the maximum waste loading for a grout. Previous work with mixtures of grout and waste has shown that unconfined compressive strength is a good indicator for a cohesive monolith and a good predictor for maximum waste loading for a grout (Loomis et al. 2003).

Data generated during the compressive-strength testing were used to support modeling of the surface barrier and contaminant transport from the treated waste form. The tests have application to ISG of:

- Transuranic (TRU) pits and trenches
- Non-TRU pits, trenches, and soil vault rows
- In situ thermal desorption (ISTD)-treated organic sludge.

Compressive strength of grouted waste forms estimates the resistance of the grouted waste form to compression in the subsurface. These data will help establish the magnitude of physical stresses required to cause fractures that could compromise the integrity surface barrier. These tests were done for ISG waste forms using nonradioactive surrogates. WAXFIX (a paraffin-based material) was added as a potential grout type to be used at the SDA. Additional tests, using WAXFIX and the various waste types, were performed. The purpose of these tests is to understand better the advantages and limitations of WAXFIX as an in situ grout for TRU and non-TRU waste, because WAXFIX has not been as extensively tested as the other grouts proposed in Yancey et al. (2003). Compressive strength was measured for waste (nonradioactive surrogates) grouted with WAXFIX.

When Yancey et al. (2003) was prepared, waste loadings were suggested for testing the WAXFIX with various waste types. When testing began, it was apparent that some of the waste loadings were not appropriate for the specific grouts. Specifically, WAXFIX was not able to handle as much organic sludge surrogate as expected but could mix with higher waste loadings of soil and nitrate salt sludge than expected. Table B-1 listed the waste loadings for compressive-strength testing that were suggested in Yancey et al. (2003) and those that were actually tested.

Table B-1. Summary of waste loadings used with WAXFIX for compressive-strength testing for in situ grouting of transuranic waste pits and trenches.

Test Method	Method/Unit	Waste Matrix	Grouts ^a	Waste in Grout Suggested in the Test Plan ^b (wt%)	Waste Loadings Used in This Test (wt%)	Replicates	Total Samples
Nonradioactive Surrogates							
ASTM C-39, D-695	Unconfined compressive strength/psi	Organic sludge surrogate	WAXFIX	10	5	5	25
				20	7		
				30	10		
				40	12		
				50	30		
ASTM C-39, D-695	Unconfined compressive strength/psi	Nitrate salt sludge surrogate	WAXFIX	10	40	5	20
				20	50		
				30	60		
				40	75		
				50			
				60			

Table B-1. (continued).

Test Method	Method/Unit	Waste Matrix	Grouts ^a	Waste in Grout Suggested in the Test Plan ^b (wt%)	Waste Loadings Used in This Test (wt%)	Replicates	Total Samples
Nonradioactive Surrogates							
ASTM C-39, D-695	Unconfined compressive strength/psi	Soil from the INL Site	WAXFIX	30	40	5	25
				40	50		
				50	60		
				60	70		
				70	80		
				80			

a. WAXFIX will contain B-10.

b. Yancey et al. 2003

In addition to the change in waste loadings used, the range of waste loadings tested was reduced for the WAXFIX samples. This was because of the inability to effectively maintain a homogeneous mixture of surrogate and WAXFIX at lower waste-grout ratios. At lower waste loadings, the surrogate and molten WAXFIX were mixed, but as soon as the mixing stopped, the surrogate would settle to the bottom before the sample could cool. This situation can be seen in Figure B-1. When these samples were tested for compressive strength, the failure always occurred at the surrogate-WAXFIX interface. The result was that there was no difference in compressive strength between samples mixed at 10, 20, 30, or 40 wt%, because failure always occurred at the waste-WAXFIX interface (see Figure B-2).

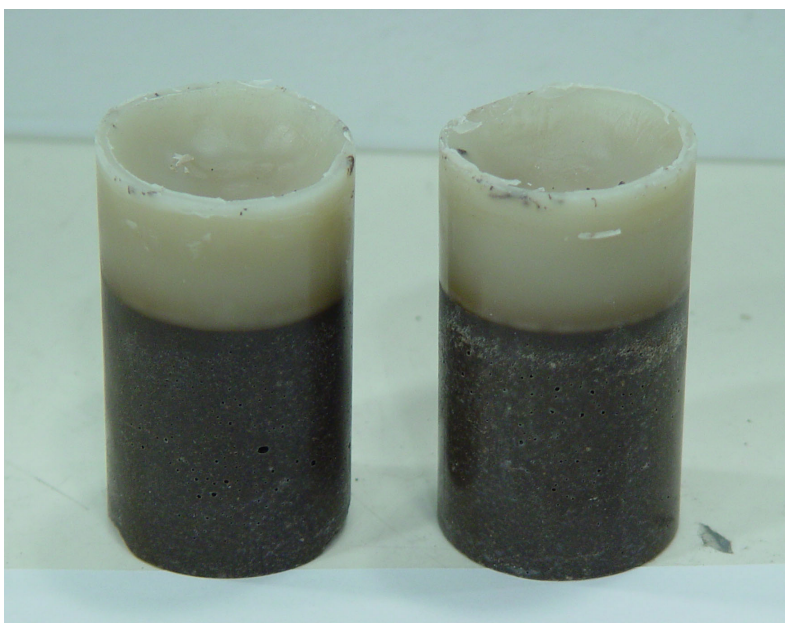


Figure B-1. WAXFIX and soil.



Figure B-2. Compressive failure of WAXFIX-soil samples.

B-2. EXPERIMENT DESIGN AND PROCEDURES

Mixtures of neat grout were prepared and allowed to cure in a 100% humidity environment for at least 30 days. WAXFIX samples did not require 100% humidity during curing and were cured at ambient humidity levels for at least 48 hours.

B-2.1 In Situ Grouting Transuranic Waste Matrix

A series of samples was prepared to determine the strength of WAXFIX grout when mixed with common waste forms found in TRU pits and trenches, including nitrate salt surrogate, organic sludge surrogate, and soil from the Idaho National Laboratory (INL) Site. Previous tests had been completed using GMENT-12, U.S. Grout, and TECT HG (Loomis et al. 2003). The samples' compatibility with typical waste matrices was measured by compressive strength (ASTM C-695).

Previously, mixtures of grout and soil from the INL Site (sieved to 10 mesh) were mixed at 12, 25, 50, and 75 wt% soil and allowed to cure in 100% humidity environment. These samples were measured by Loomis et al. (2003). During this study, only WAXFIX was tested with soil from the INL Site at waste loadings of 40, 50, 60, 70, and 80 wt% soil. This deviated from Yancey et al. (2003) ratios of 10, 20, 30, 40, 50, and 60 wt% because, during mixing of samples, it was discovered that WAXFIX could handle higher waste loadings than previously thought. The waste loadings tested, therefore, were increased to match the capability of WAXFIX. The waste loading that first showed a marked decrease was noted for comparison with other grouts to determine waste loadings for other tests such as hydraulic conductivity.

B-2.2 In Situ Grouting of ISTD-Treated Transuranic Waste Matrix

Compressive strength was tested with varying ratios of grout and thermally desorbed (450°C [842°F]), nonradioactive, surrogate organic sludge to determine how much ash can be mixed with grout to form a freestanding monolith having greater than 250 psi compressive strength.

Mixture compositions starting at 5 wt% with increasing intervals were tested for compressive strength, until the compressive strength fell to less than 250 psi for each of the four grouts (see Table B-2). The grout and thermally desorbed material was mixed, poured into 2-in.-diameter by 4-in.-high cylindrical molds, and allowed to cure in 100% relative humidity for at least 30 days. Compressive strength was tested according to ASTM C39, using five samples per waste-loading range.

Table B-2. Summary of compressive-strength testing for in situ grouting of waste from ISTD-treated transuranic waste pits and trenches.

Test Method	Method/Unit	Waste Matrix	Analytes	Grouts ^a	Waste in Grout (wt%)	Replicates	Total Samples
Nonradioactive Surrogate							
ASTM C-39, D-695	Unconfined compressive strength/psi	ISTD-treated organic sludge surrogate	N/A	GMMENT-12, U.S. Grout, TECT HG, WAXFIX	5 10 15 20 30 50	5	120

a. WAXFIX contains B-10.

B-2.3 In Situ Grouting of Nontransuranic Waste Matrix

The soil from the INL Site was used to simulate waste in the non-TRU pits and trenches and soil vault rows. Previous tests by Loomis et al. (2003) had been completed using GMMENT-12, U.S. Grout, and TECT HG. The soil from the INL Site was used under the TRU waste matrix to simulate waste expected in the TRU pits and trenches waste. The soil from the INL Site also was used as a surrogate for non-TRU waste. Since both TRU and non-TRU tests required soil from the INL Site as a surrogate, the test was performed only once, but the data were used both for TRU and non-TRU waste.

B-3. EQUIPMENT AND MATERIALS

Nonradioactive surrogate material was prepared for mixing with nitrate salt sludge and organic sludge. The recipes for these surrogates are found in Appendix A (see Tables B-3 and B-4). Samples were prepared using 2-in.-diameter by 4-in.-tall plastic molds. Compressive strength was tested using an Instron 4505 screw-driven load frame with a 22.48-kip load cell (see Figure B-3). Data were collected electronically using LabVIEW as the data acquisition platform.

Table B-3. Subsurface Disposal Area organic sludge (Rocky Flats Plant Series 743) for ISTD and in situ grouting testing.

Material	Specific Gravity ^a	Historic Estimated Concentration				Selected Concentration (wt%)
		(vol%)		(wt%)		
		1981 ^b	1998 ^c	1981	1998 ^c	
Texaco Regal oil	0.87	22.0	16	20	14	29.0
Miscellaneous oil ^d	0.90	11.0 ^d	8	13	10	—
Carbon tetrachloride	1.59	17.0	19	27	30	27.0
TCE ^c	1.46	4.0	5	6	6	7.0

Table B-3. (continued).

Material	Specific Gravity ^a	Historic Estimated Concentration				Selected Concentration (wt%)
		(vol%)		(wt%)		
		1981 ^b	1998 ^c	1981	1998 ^c	
TCA	1.44	5.0	6	9	8	9.0
PCE ^d	1.59	4.0	5	6	6	7.0
Silicon oxide	2.50	8.4	12	21	29	13.5
Oil Dri	2.30	—	—	—	—	7.3

a. The organic chlorinated solvents are over 60% denser than the oils (specific gravity is 1.5 versus 0.87).

b. The year of records and estimation.

c. (Miller and Navratil 1998).

d. 43% miscellaneous oil (mineral oil used for the surrogate) and solvents listed in shipping records, of which 20% has been divided between TCE and PCE. The TCE and PCE were put here from the miscellaneous oil category.

PCE = tetrachloroethene

TCA = trichloroethane

TCE = trichloroethene

Table B-4. Subsurface Disposal Area nitrate salt sludge (Rocky Flats Plant Series 745) for in situ grouting and ISTD testing.

Material	Historical Content (wt%)		Bench Test Batch Using Typical Salts (wt%)	
	1978	1992		
Sodium	16.20	23.90	Sodium nitrite	0.50
Potassium	14.50	8.60	Potassium nitrate	30.00
Nitrate	62.30	54.90	Sodium nitrate	60.00
Chlorine	2.80	3.10	Sodium chloride	3.00
Fluorine	0.30	0.60	Sodium fluoride	0.50
Phosphate	1.30	1.40	Sodium phosphate	1.00
Sulfate	3.60	3.60	Sodium sulfate	3.00
Carbonate	N/A	0.40	Sodium bicarbonate	1.00
Chromium	0.03	0.04	Chromium nitrate	0.04
Organics	N/A	1.00	EDTA	1.00
Water	N/A	2.00	Water	2.00
Total	99.10	99.50	Water added to above mix 9(%)	100.00

EDTA = ethylene diaminetetraacetic

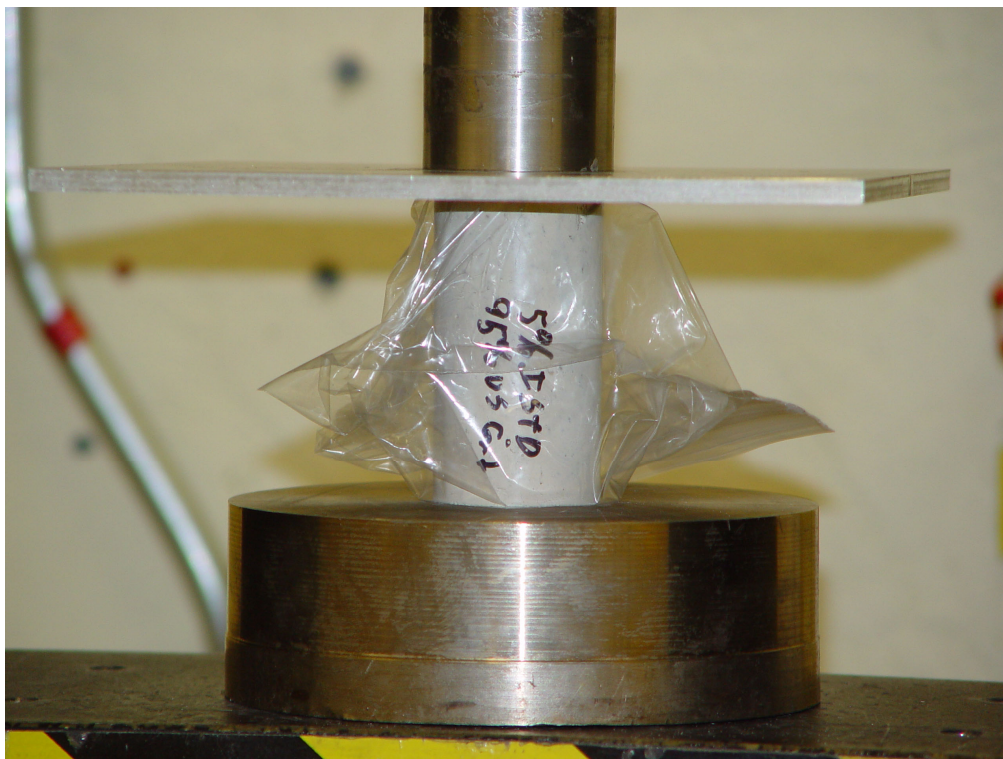


Figure B-3. Compressive-strength grouted waste using an Instron 4505 screw-driven load frame with a 22.48-kip load cell.

B-4. DATA MANAGEMENT, ANALYSIS, AND INTERPRETATION

Compressive-strength data were measured in pounds per square inch (psi), and samples were measured in replicates of five. Averages were determined and error bars presented on figures using the 95% confidence interval. The physical property data from these tests are used to predict the long-term physical stability of the grouted waste forms. The test results also support the risk assessment, risk model, and performance evaluation portions of the Operable Unit 7-13/14 feasibility study.

B-5. RESULTS AND CONCLUSIONS

The results of the compressive-strength testing are presented in four categories: neat grout, ISG of TRU waste, ISG of ISTD-treated TRU waste, and ISG of Non-TRU waste. The neat grout was tested to establish a basis of comparison for the grouts.

B-5.1 Neat Grout

Compressive strength was measured for all of the grouts without adding the surrogates in order to establish a baseline for the neat grouts. The results shown in Table B-5 show that the cementitious grouts demonstrate much higher compressive strength than that observed in the WAXFIX. In this study, the compressive strength of the neat grouts shows GMENT-12 having the highest compressive strength, followed by TECT HG, and then U.S. Grout. All of the cementitious grouts had compressive-strength values well above the 250-psi minimum criterion established for this test. WAXFIX compressive strength also was higher than 250 psi but was an order of magnitude lower than the cementitious grouts.

Table B-5. Compressive-strength values for the neat grouts.

Test	Grout Product			
	WAXFIX	GMENT-12	TECT HG	U.S. Grout
Compressive strength, Specimen A (psi)	276	4,418	3,180	1,546
Compressive strength, Specimen B (psi)	295	5,160	2,786	2,220
Compressive strength, Specimen C (psi)	304	5,719	2,879	2,001
Compressive strength, Specimen D (psi)	324	5,200	2,907	1,283
Compressive strength, Specimen E (psi)	296	5,674		1,305
Average compressive strength (psi)	299	5,234	2,934	1,674
Standard deviation	17	525	175	421
95% confidence interval	21	651	278	523

B-5.2 In Situ Grouting of Transuranic Waste

The waste matrices used to simulate those expected in the TRU waste include soil, organic sludge, and nitrate salt sludge. This section presents the result for compressive strength for each of the grout types and the waste matrix being tested for TRU waste surrogates.

B-5.3 Soil as an Interference

Considerable tolerance to soil loading was observed with all of the grouts. GMENT-12, TECT HG, U.S. Grout, and WAXFIX all pass testing for soil tolerance. The data for GMENT-12, TECT HG, and U.S. Grout were taken from Loomis et al. (2003). GMENT-12 had the highest compressive strength, while WAXFIX, as expected, showed the lowest compressive strength. Table B-6 shows the test results for grout and soil of 12, 25, 50, and 75 wt% for the cementitious grouts and 40, 50, 60, 70, and 80 wt% for WAXFIX. GMENT-12 had the highest compressive strength at the 50 wt% soil loadings, greater than 2,500 psi (see Figure B-4). The error bars in Figure B-4 show the 95% confidence interval for the data tested. Differences observed with the averages and error bars are statistically different with 95% confidence. Adding soil to grout resulted in a higher average compressive strength for 25 wt% than for neat grout, much the same way that adding aggregate to concrete in the building industry strengthens the concrete. The data support a recommendation for using 50 wt% soil during the physical and chemical testing for cementitious grouts and that 70 wt% waste loadings be used on subsequent physical and chemical tests for WAXFIX.

Table B-6. Individual compressive-strength test results for specimens containing soil from the Idaho National Laboratory Site interference at various loadings.

Specimen	Waste Type	Compressive Strength of Grout (psi)				U.S. Grout ^a
		Waste Loading (wt%)	WAXFIX	GMENT-12 ^a	TECT HG ^a	
Specimen A	Soil from the INL Site	12	—	5,734	3,759	3,98
Specimen B	Soil from the INL Site	12	—	5,145	4,227	3,80
Specimen C	Soil from the INL Site	12	—	6,774	4,464	3,90
	Mean	—	—	5,884	4,150	3,89
	Standard Deviation	—	—	825	359	8
	95% Confidence Interval	—	—	1,024	445	11
Specimen A	Soil from the INL Site	25	—	5,876	3,501	2,99
Specimen B	Soil from the INL Site	25	—	5,855	3,762	3,15
Specimen C	Soil from the INL Site	25	—	6,413	3,698	3,13
	Mean	—	—	6,048	3,654	3,09
	Standard deviation	—	—	316	136	8
	95% confidence interval	—	—	393	169	11
Specimen A	Soil from the INL Site	40	350	—	—	—
Specimen B	Soil from the INL Site	40	441	—	—	—
Specimen C	Soil from the INL Site	40	403	—	—	—
Specimen D	Soil from the INL Site	40	366	—	—	—
Specimen E	Soil from the INL Site	40	270	—	—	—
	Mean	—	366	—	—	—
	Standard deviation	—	53	—	—	—
	95% confidence interval	—	66	—	—	—
Specimen A	Soil from the INL Site	50	228	2,722	1,884	1,18
Specimen B	Soil from the INL Site	50	206	2,263	1,927	1,42
Specimen C	Soil from the INL Site	50	198	2,602	1,962	1,22
Specimen D	Soil from the INL Site	50	204	—	—	—
Specimen E	Soil from the INL Site	50	225	—	—	—
	Mean	—	212	2,529	1,924	1,27
	Standard deviation	—	13	238	39	12
	95% confidence interval	—	17	591	97	31
Specimen A	Soil from the INL Site	60	303	—	—	—
Specimen B	Soil from the INL Site	60	315	—	—	—
Specimen C	Soil from the INL Site	60	356	—	—	—
Specimen D	Soil from the INL Site	60	367	—	—	—
Specimen E	Soil from the INL Site	60	391	—	—	—
	Mean	—	346	—	—	—
	Standard deviation	—	37	—	—	—
	95% confidence interval	—	45	—	—	—

Table B-6. (continued).

Specimen	Waste Type	Compressive Strength of Grout (psi)				
		Waste Loading (wt%)	WAXFIX	GMENT-12 ^a	TECT HG ^a	U.S. Grout ^a
Specimen A	Soil from the INL Site	70	628	—	—	—
Specimen B	Soil from the INL Site	70	696	—	—	—
Specimen C	Soil from the INL Site	70	790	—	—	—
Specimen D	Soil from the INL Site	70	659	—	—	—
Specimen E	Soil from the INL Site	70	584	—	—	—
	Mean	—	671	—	—	—
	Standard deviation	—	78	—	—	—
	95% confidence interval	—	97	—	—	—
Specimen A	Soil from the INL Site	75	—	—	—	75
Specimen B	Soil from the INL Site	75	—	—	—	83
Specimen C	Soil from the INL Site	75	—	—	—	82
	Mean	—	—	—	—	80
	Standard deviation	—	—	—	—	4
	95% confidence interval	—	—	—	—	10
Specimen A	Soil from the INL Site	80	367	—	—	—
Specimen B	Soil from the INL Site	80	355	—	—	—
Specimen C	Soil from the INL Site	80	292	—	—	—
Specimen D	Soil from the INL Site	80	270	—	—	—
Specimen E	Soil from the INL Site	80	358	—	—	—
	Mean	—	328	—	—	—
	Standard deviation	—	44	—	—	—
	95% confidence interval	—	55	—	—	—

a. The results for GMENT-12, TECT HG, and U.S. Grout were taken from Loomis et al. (2003).

B-5.4 Organic Sludge as an Interference

Organic sludge, when mixed with neat grout during the jet grouting process, has the potential to produce zones of considerably degraded grout (increased hydraulic conductivity and loss of compressive strength). In TRU pits and trenches, organic sludge makes up an average of 5 vol% of the waste (Loomis et al. 2003); however, zones of almost total drums of organic sludge are possible. A study (Loomis, Zdinak, and Bishop 1997) shows that jet grouting into greaselike materials can degrade grout curing and monolith stability; however, when isolated drums of organic material are jet grouted, a cohesive monolith can be achieved. Grout was mixed with an organic sludge surrogate formulation based on Rocky Flats Plant waste (see Table B-3) using trichlorethylene, tetrachloroethylene, carbon tetrachloride, and trichloroethane as volatile organics mixed with absorbents and Texaco Regal Oil. The resultant mixture of volatile organics, oil, and absorbents exhibited a greaselike consistency like that expected with TRU organic sludge waste.

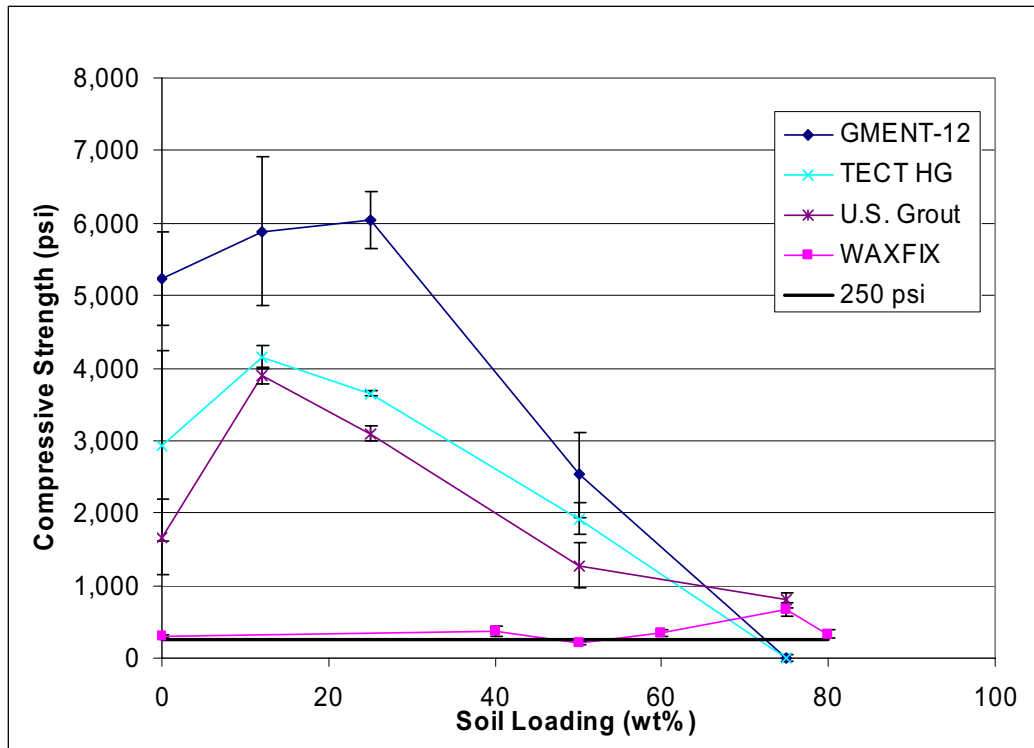


Figure B-4. Compressive strength of soil and grout mixtures.

MENT-12, TECT HG, and U.S. Grout demonstrated good tolerance to organic content for lower loadings of the organic sludge (up to 12 wt%), as shown on Table B-7. However, for loadings higher than 12-wt% organic sludge, the resultant monoliths exhibited poor compressive strength. Figure B-5 summarizes the individual test results, showing that GMENT-12 had very little degradation and in fact maintained a relatively high compressive strength (nominally 6,000 psi) for all triplicate samples through 9-wt% organic sludge. The GMENT-12 showed significantly higher compressive strength up to 12 wt% than the other grouts at the 95% confidence interval. TECT HG grout also had reasonably high compressive strength (3,000-4,000 psi) for up to 12 wt% and even tolerated 25 wt% sludge at an average compressive strength of 2,347 psi, which is consistent with samples obtained during past grouting experiments (Loomis, Zdinak, and Bishop 1997). Based on the results shown in Table B-7, physical and chemical testing for grouted interferences (discussed in a following section) should be performed at 9 wt% for cementitious grouts and 10 wt% loading for WAXFIX.

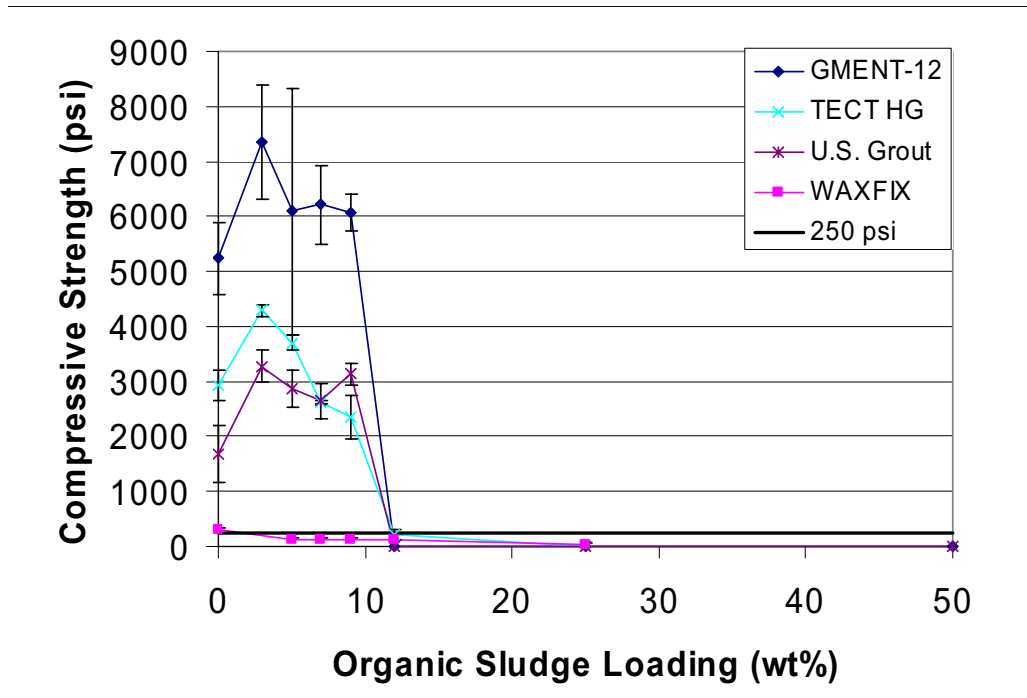


Figure B-5. Compressive strength of organic sludge and grout mixtures.

Table B-7. Results in pound per square inch of individual compressive-strength tests for interference tolerance of specimens containing the organic sludge interference at various loadings.

Specimen	Interference Type	Interference Percentage	Grout Product			
			WAXFIX	MENT-12	TECT HG	U.S. Grout
Specimen A	Organic sludge	3	—	7,460	4,230.0	3,202.0
Specimen B	Organic sludge	3	—	6,456	4,266.0	3,084.0
Specimen C	Organic sludge	3	—	8,131	4,391.0	3,542.0
Mean		—	—	7,349	4,295.7	3,276.0
Standard deviation		—	—	843	85.0	238.0
95% confidence interval		—	—	1,047	105.0	295.0
Specimen A	Organic sludge	5	131.0	5,077	3,764.0	3,010.0
Specimen B	Organic sludge	5	125.0	6,788	3,664.0	2,736.0
Specimen C	Organic sludge	5	136.0	6,434	3,690.0	2,887.0
Specimen D	Organic sludge	5	103.0	—	—	—
Specimen E	Organic sludge	5	121.0	—	—	—
Mean		—	123.0	6,100	3,706.0	2,878.0
Standard deviation		—	13	903	52.0	137.0
95% confidence interval		—	16	2,244	129.0	341.0
Specimen A	Organic sludge	7	140.0	6,463	2,805.0	2,501.0
Specimen B	Organic sludge	7	121.0	5,897	2,827.0	2,746.0
Specimen C	Organic sludge	7	107.0	6,286	2,828.0	2,685.0

Table B-7. (continued).

Specimen	Interference Type	Grout Product				
		Interference Percentage	WAXFIX	GMENT-12	TECT HG	U.S. Grout
Specimen D	Organic sludge	7	130.0	—	—	—
Specimen E	Organic sludge	7	122.0	—	—	—
	Mean	—	124.0	6,215	2,820.0	2,644.0
	Standard deviation	—	12.0	290	13.0	128.0
	95% confidence interval	—	15.0	719	32.0	317.0
Specimen A	Organic sludge	9	—	6,123	2,586.0	3,161.0
Specimen B	Organic sludge	9	—	6,194	2,650.0	3,047.0
Specimen C	Organic sludge	9	—	5,932	—	3,201.0
	Mean	—	—	6,083	2,618.0	3,136.3
	Standard deviation	—	—	136	45.0	80.0
	95% confidence interval	—	—	337	407.0	199.0
Specimen A	Organic sludge	10	125.0	—	—	—
Specimen B	Organic sludge	10	66.0	—	—	—
Specimen C	Organic sludge	10	132.0	—	—	—
Specimen D	Organic sludge	10	110.0	—	—	—
Specimen E	Organic sludge	10	127.0	—	—	—
	Mean	—	112.0	—	—	—
	Standard deviation	—	27.0	—	—	—
	95% confidence interval	—	33.0	—	—	—
Specimen A	Organic sludge	12	105.0	—	2,349.0	—
Specimen B	Organic sludge	12	105.0	—	2,308.0	—
Specimen C	Organic sludge	12	129.0	—	2,383.0	—
Specimen D	Organic sludge	12	113.0	—	—	—
Specimen E	Organic sludge	12	120.0	—	—	—
	Mean	—	114.0	—	2,347.0	—
	Standard deviation	—	10.0	—	38.0	—
	95% confidence interval	—	13.0	—	93.0	—
Specimen A	Organic sludge	25	—	—	204.0	—
Specimen B	Organic sludge	25	—	—	—	—
Specimen C	Organic sludge	25	—	—	—	—
	Mean	—	—	—	204.0	—
	Standard deviation	—	—	—	—	—
	95% confidence interval	—	—	—	—	—
Specimen A	Organic sludge	30	47.9	—	—	—
Specimen B	Organic sludge	30	53.7	—	—	—
Specimen C	Organic sludge	30	31.6	—	—	—
Specimen D	Organic sludge	30	45.1	—	—	—
Specimen E	Organic sludge	30	47.8	—	—	—

Table B-7. (continued).

Specimen	Interference Type	Grout Product				
		Interference Percentage	WAXFIX	GMMENT-12	TECT HG	U.S. Grout
	Mean	—	45.0	—	—	—
	Standard deviation	—	8.0	—	—	—
	95% confidence interval	—	10.0	—	—	—
Specimen A	Organic sludge	50	—	—	6.0	—
Specimen B	Organic sludge	50	—	—	7.0	—
Specimen C	Organic sludge	50	—	—	—	—
	Mean	50	—	—	6.5	—
	Standard deviation	—	—	—	0.71	—
	95% confidence interval	—	—	—	6	—

B-5.5 Nitrate Salt as an Interference

Each of the grouts was mixed with nitrate salts in granular form (roughly 33% potassium nitrate and 67% sodium nitrate, representing Rocky Flats Plant evaporation pond salts found in the TRU pits and trenches in the SDA at the INL Site) at various waste loadings (12, 25, 50, and 75 wt%). Salts in general have been shown to cause degradation of concretes; therefore, knowledge of the grout tolerance of each grout to these nitrate salts is important for determining long-term integrity of the monoliths.

Following curing, compressive strength of the monoliths was tested in sets of five. Results are presented in Table B-8. U.S. Overall, there was a significant amount of variability between the compressive strengths of the grouts tested. As a result, there were no differences observed between the grouts at the 95% confidence level. However, at the 50 wt% loading of nitrate salt sludge surrogate, the U.S. Grout demonstrated a compressive strength significantly higher than the other grouts. Based on the results shown in Table B-8, a nitrate loading of 12 wt% should be used on cementitious grout to test physical and chemical interference of nitrate salts. Compressive strength of WAXFIX and nitrate salt sludge remained relatively constant up to 60 wt% waste loading (see Figure B-6) and was significantly lower than the other grouts tested.

Table B-8. Individual compressive-strength test results in pound per square inch for the interference tolerance testing of specimens containing the nitrate salt interference at various loadings.

Interference					
Specimen	(%)	WAXFIX	GMMENT-12	TECT HG	U.S. Grout
Specimen A	12	—	1,906	3,224	5,298
Specimen B	12	—	2,906	3,254	4,617
Specimen C	12	—	4,702	—	4,490
Mean	—	—	3,171	3,239	4,802
Standard deviation	—	—	1,417	21	435
95% confidence interval	—	—	3,520	191	1,079
Specimen A	25	—	2,948	1,198	1,306
Specimen B	25	—	2,298	1,184	1,420
Specimen C	25	—	3,408	1,196	1,423

Table B-8. (continued).

Specimen	Interference (%)	WAXFIX	GMMENT-12	TECT HG	U.S. Grout
Mean	—	—	2,883	1,193	1,383
Standard deviation	—	—	558	8	67
95% confidence interval	—	—	1,386	19	166
Specimen A	40	171	—	—	—
Specimen B	40	220	—	—	—
Specimen C	40	222	—	—	—
Specimen D	40	228	—	—	—
Specimen E	40	231	—	—	—
Mean	—	215	—	—	—
Standard deviation	—	25	—	—	—
95% confidence interval	—	30	—	—	—
Specimen A	50	184	3	—	1,819
Specimen B	50	188	3	—	1,765
Specimen C	50	185	2	—	1,857
Specimen D	50	182	—	—	—
Specimen E	50	186	—	—	—
Mean	—	185	2.7	—	1,813
Standard deviation	—	2.6	0.58	—	46
95% confidence interval	—	3.2	1.43	—	115
Specimen A	60	244	—	—	—
Specimen B	60	179	—	—	—
Specimen C	60	202	—	—	—
Specimen D	60	173	—	—	—
Specimen E	60	165	—	—	—
Mean	—	192	—	—	—
Standard deviation	—	32	—	—	—
95% confidence interval	—	40	—	—	—
Specimen A	75	<100	—	—	873
Specimen B	75	<100	—	—	866
Specimen C	75	<100	—	—	868
Specimen D	—	<100	—	—	—
Specimen E	—	<100	—	—	—
Mean	—	—	104	—	869
Standard deviation	—	—	7.8	—	3.6
95% confidence interval	—	—	19	—	9.0

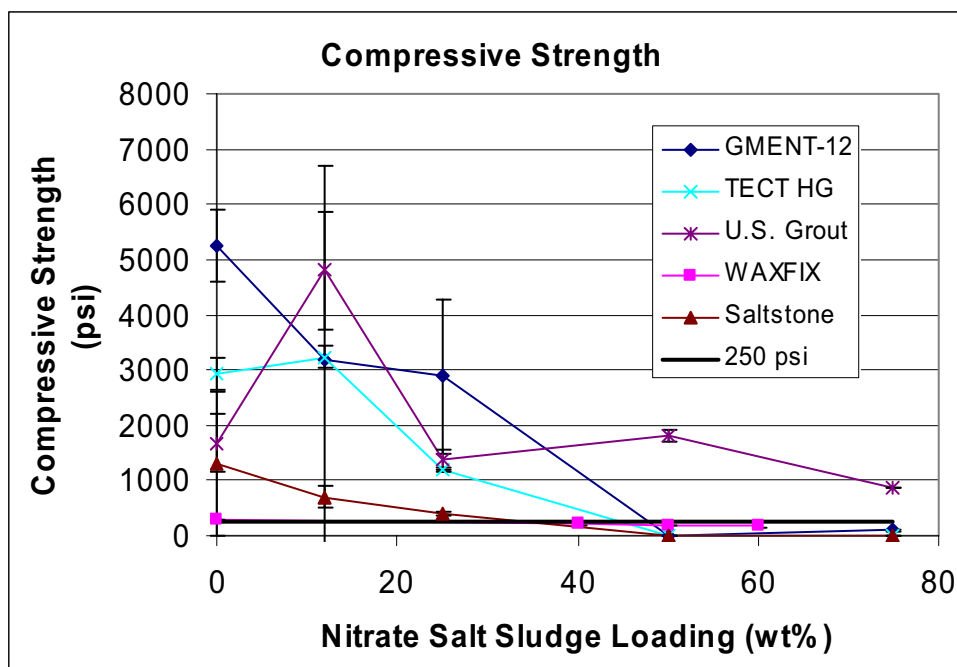


Figure B-6. Compressive strength with nitrate salt sludge as a surrogate.

As with soil, achieving a constant consistent mixture of hot and molten WAXFIX and nitrate salt sludge was difficult. As soon as mixing stopped, the salts would settle to the bottom of the sample, leaving nearly neat WAXFIX on top with a high loading of salt in the wax on the bottom (see Figure B-7). The loading of 60 wt% was found to allow for complete mixing with the wax with minimal separation.

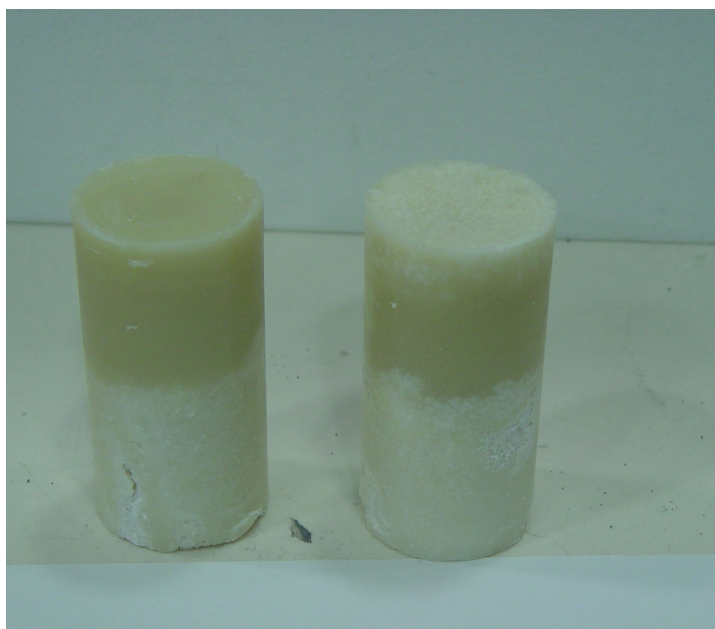


Figure B-7. WAXFIX and nitrate salt sludge surrogate.

B-5.6 ISTD-Treated Organic Sludge as an Interference

Neat grouts were mixed with thermally desorbed organic sludge at various loadings (12, 25, 50, and 75 wt%). Mixing grouts with the thermally treated organic sludge, as expected, resulted in higher waste loading than grout mixed with untreated organic sludge. Thermally treating the organic sludge dried the waste form significantly, which resulted in need for adding extra water to all of the grouts to get good mixing at the 30- and 50-wt% waste loadings.

Following curing, compressive strength of the monoliths was tested in sets of five. The results are presented in Table B-9. All of the grouts performed well, even at 50-wt% waste loading. However, 30-wt% loading was selected for subsequent chemical and physical tests, because it was easier to mix and prepare. GMENT-12 showed the best tolerance to thermally treated organic sludge loadings with compressive strength in excess of 2,500 psi at 30 wt% (see Figure B-8). Based on the results shown in Table B-9, a waste loading of 30 wt% was recommended as the waste loading to use during physical and chemical testing of interference caused by thermally treated organic sludge.

Table B-9. Results in pound per square inch of individual compressive-strength tests for each grout mixed with ISTD-treated organic sludge.

Specimen	Interference Type	Interference (%)	Grout Product			
			WAXFIX	GMMENT-12	TECT HG	U.S. Grout
Specimen A	ISTD-treated organic sludge	5	—	2,489	—	1,207
Specimen B	ISTD-treated organic sludge	5	—	3,554	—	1,317
Specimen C	ISTD-treated organic sludge	5	—	2,675	—	1,180
Specimen D	ISTD-treated organic sludge	5	—	2,388	—	2,201
Specimen E	ISTD-treated organic sludge	5	—	2,840	—	1,913
	Mean	—	—	2,789.2	—	1,563.6
	Standard deviation	—	—	461	—	465
	95% confidence interval	—	—	573	—	577
Specimen A	ISTD-treated organic sludge	10	—	2,367	2,275	1,419
Specimen B	ISTD-treated organic sludge	10	—	2,697	1,563	1,520
Specimen C	ISTD-treated organic sludge	10	—	2,905	2,010	1,743
Specimen D	ISTD-treated organic sludge	10	—	3,043	1,579	1,840
Specimen E	ISTD-treated organic sludge	10	—	2,733	—	—
	Mean	—	—	2,749	1,857	1,631
	Standard deviation	—	—	255	347	194
	95% confidence interval	—	—	316	553	309
Specimen A	ISTD-treated organic sludge	15	—	2,766	—	—
Specimen B	ISTD-treated organic sludge	15	—	2,619	—	—
Specimen C	ISTD-treated organic sludge	15	—	2,652	—	—
Specimen D	ISTD-treated organic sludge	15	—	2,727	—	—
Specimen E	ISTD-treated organic sludge	15	—	2,713	—	—
	Mean	—	—	2,696	—	—
	Standard deviation	—	—	59	—	—
	95% confidence interval	—	—	74	—	—

Table B-9. (continued).

Specimen	Interference Type	Interference (%)	Grout Product			
			WAXFIX	GMMENT-12	TECT HG	U.S. Grout
Specimen A	ISTD-treated organic sludge	20	—	2,161	1,932	2,221
Specimen B	ISTD-treated organic sludge	20	—	2,396	2,095	1,695
Specimen C	ISTD-treated organic sludge	20	—	2,391	2,580	2,438
Specimen D	ISTD-treated organic sludge	20	—	3,086	1,976	2,118
Specimen E	ISTD-treated organic sludge	20	—	3,111	—	2,110
	Mean	—	—	2,629	2,116.4	2,146
	Standard deviation	—	—	439	298	298
	95% confidence interval	—	—	545	473	473
Specimen A	ISTD-treated organic sludge	30	269	2,020	2,200	2,326
Specimen B	ISTD-treated organic sludge	30	263	3,441	1,961	1,668
Specimen C	ISTD-treated organic sludge	30	133	2,356	2,073	1,740
Specimen D	ISTD-treated organic sludge	30	186	2,835	—	2,217
Specimen E	ISTD-treated organic sludge	30	170	2,217	—	1,375
	Mean	—	204.2	2,574	1,865.2	2,078
	Standard deviation	—	60	570	120	120
	95% confidence interval	—	74	708	297	297
Specimen A	ISTD-treated organic sludge	40	240	—	—	—
Specimen B	ISTD-treated organic sludge	40	248	—	—	—
Specimen C	ISTD-treated organic sludge	40	263	—	—	—
Specimen D	ISTD-treated organic sludge	40	194	—	—	—
Specimen E	ISTD-treated organic sludge	40	238	—	—	—
	Mean	—	237	—	—	—
	Standard deviation	—	26	—	—	—
	95% confidence interval	—	32	—	—	—
Specimen A	ISTD-treated organic sludge	50	227	1,198	931	890
Specimen B	ISTD-treated organic sludge	50	234	1,692	976	716
Specimen C	ISTD-treated organic sludge	50	229	1,688	660	1,361
Specimen D	ISTD-treated organic sludge	50	227	1,360	696	657
Specimen E	ISTD-treated organic sludge	50	245	1,050	1,143	640
	Mean	—	232	1,398	852.8	881
	Standard deviation	—	7	288	202	202
	95% confidence interval	—	9	358	251	251
Specimen A	ISTD-treated organic sludge	60	227	—	—	—
Specimen B	ISTD-treated organic sludge	60	240	—	—	—
Specimen C	ISTD-treated organic sludge	60	239	—	—	—
Specimen D	ISTD-treated organic sludge	60	270	—	—	—
Specimen E	ISTD-treated organic sludge	60	242	—	—	—
	Mean	—	243	—	—	—
	Standard deviation	—	16	—	—	—
	95% confidence interval	—	20	—	—	—

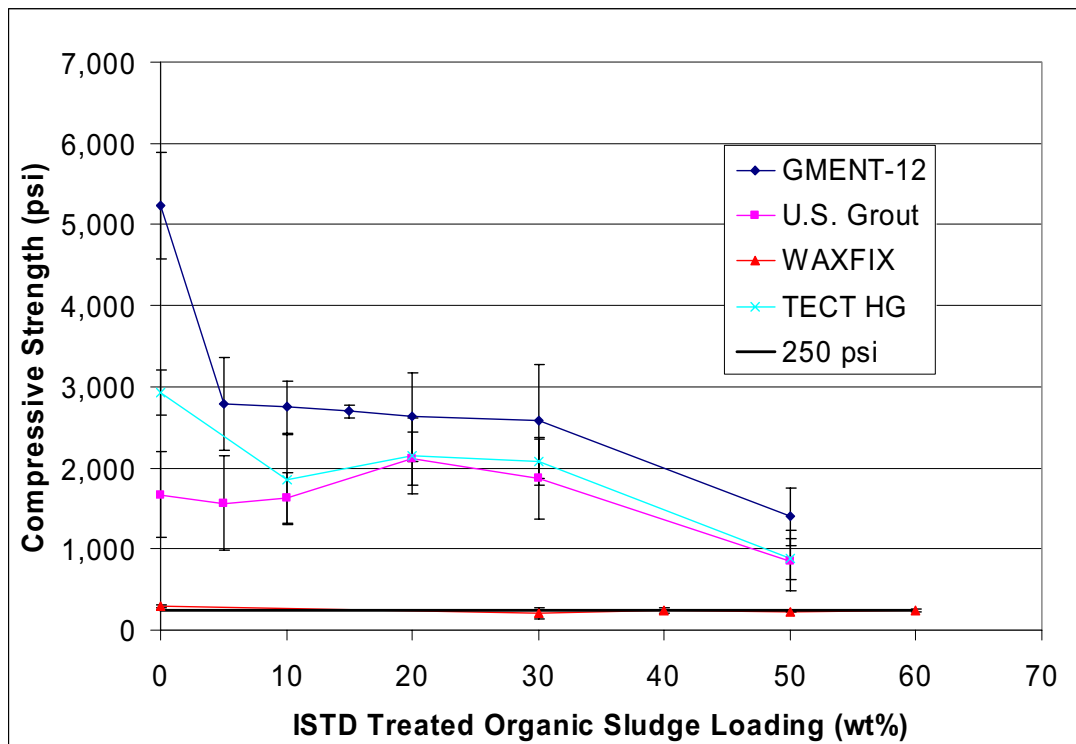


Figure B-8. Compressive strength with ISTD-treated organic sludge as a surrogate.

B-5.7 In Situ Grouting of Nontransuranic Waste

The surrogate used for non-TRU waste in this study was soil. Rather than duplicate the test performed for the TRU waste samples, the results listed under the TRU section using soil as the surrogate were used to support this section. Please refer to Section B5.3.

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Appendix C

Porosity Tests—Part 1

CONTENTS

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Appendix C

Porosity Tests—Part 1

Porosity is used in mechanistic modeling of release from treated waste and is a fundamental modeling input that controls both the interstitial velocity at which a given Darcian water flux moves and the diffusive fluxes in both the aqueous and gaseous phases. The porosity of a porous medium is the ratio of the volume of void spaces to the total sample volume.

To determine the volume of the void spaces in a porous medium, the voids must be filled completely with a fluid that can be measured. By flushing the samples with carbon dioxide gas and then saturating the samples with de-aerated water under a vacuum, the pores can be filled with water in the absence of entrapped air, allowing calculation of sample porosity (Dane and Topp 2002). The ASTM standard for saturating samples to determine hydraulic conductivity (ASTM D2434-68) and methods described in Dane and Topp (2002) were referenced for direct measurements of porosity. In consolidated porous media, measured porosity is a measurement of the void space of interconnected pores. Dead-end pores are those with no connection either to other pores or to the outside of the sample. These dead-end pores will remain void of water even during saturation. Dead-end pores are not included in this measurement since water cannot be imbibed into the pores during sample saturation.

Estimated or calculated porosity from sample dry bulk density measurements includes the void space of all pores in the sample (Jury, Gardner, and Gardner 1991). Differences between calculated and measured porosity may be because of uncertainties in material or particle density values or because of dead-end pores present in the sample.

C-1. TEST OBJECTIVES, RATIONALE, AND DEVIATIONS FROM TEST PLAN

In Appendix B, the compressive strength for four grouts mixed with four surrogates was measured at varying ratios of grout to waste. One of the purposes for the compressive strength testing was to determine a maximum waste loading for a given grout-waste mixture where the waste form would maintain a freestanding monolith. We used this maximum waste loading to prepare the grout-waste mixtures in both the porosity tests presented in this section and the hydraulic conductivity tests presented in Appendix D. By testing the porosity of the neat grout and the porosity of the grout-treated waste forms at the maximum loading, the upper and lower porosity values can be determined for in situ conditions. The maximum loadings determined in Section 3.1 for mixtures of the grouts tested with soil from the Idaho National Laboratory (INL) Site were 50 wt% waste loading for GMENT-12, TECT HG, and U.S. Grout and 70 wt% waste loading for WAXFIX mixtures with soil from the INL Site.

Results obtained from these tests will be used to compare porosities between the neat grout and the grout-treated soil from the INL Site and to evaluate long-term physical stability of the grout-treated waste forms. Porosity was estimated for in-situ-grouting-treated waste in Operable Unit 7-13/14 from data generated using uncontaminated soil from the INL Site. However, because dead-end pores do not contribute to fluid transport, direct measurements of matrix porosity provide more realistic values for discerning the potential for water transport through the grout-treated waste forms than estimates based on sample dry bulk density.

Porosity measurements were made on both neat grout samples and on grout-treated waste mixtures of 50 wt% waste loading (see Table C-1). This was consistent with Yancey et al. (2003) with one

exception—WAXFIX was tested as neat grout and as a grout-treated mixture of 70 wt% waste loading rather than 50 wt% loading. The waste loading was originally determined to be 50 wt% based on the maximum loading necessary to maintain a cohesive sample for the Portland-cement-based grouts; however, the WAXFIX-INL Site soil mixtures could handle a 70 wt% loading rather than a 50 wt% loading and still maintain a cohesive sample.

Table C-1. Summary of porosity testing for nontransuranic pits and trenches and soil vault row waste.

Method	Waste Matrix	Grouts	Waste in Grout (wt%)	Replicates	Total Samples
Porosity test for aggregate concrete (ASTM D2434-68)	Soil from the INL Site	WAXFIX	0 70	3	6
Porosity test for aggregate concrete (ASTM D2434-68)	Soil from the INL Site	GMENT-12, U.S. Grout, and TECT HG	0 50	6	36

Yancey et al. (2003) stated that a minimum of three samples for each sample type should be tested to determine measurement precision. Six replicates were prepared for the GMENT-12, TECT HG, and U.S. Grout waste mixtures just in case some of the samples cracked or were destroyed in the testing process. Since none of the samples were destroyed in the sample preparation process, all six samples were tested for the cement-based grouts.

C-2. EXPERIMENTAL DESIGN AND PROCEDURES

The grouted samples were prepared using 25-mm (1-in.)-diameter by 45-mm (1.8-in.)-tall Delrin plastic sample holders, as shown in Figure C-1. Samples made with GMENT-12 and U.S. Grout were allowed to cure 30 days under 98–99% humidity in a humidity chamber. TECT HG samples were cured for 30 days in a sealed container at 100% humidity. WAXFIX samples were cured for no less than 48 hours. Curing times and conditions were established by manufacturer specification. The water used in the porosity measurements was an INL Site groundwater surrogate (see Appendix B).



Figure C-1. Porosity samples.

C-3. EQUIPMENT AND MATERIALS

Materials used to saturate the samples include a vacuum dessicator, a 600-mm (23.6-in.) mercury vacuum pump, a carbon dioxide tank, INL Site synthetic groundwater (see Appendix B), and a water de-aerator (see Figure C-2). The INL Site synthetic groundwater was de-aerated by autoclaving the water in a 7.6-L (2-gal) carboy at 121°C (249.8°F) and 21 psi for 25 minutes. The water in the carboy was allowed to cool to room temperature while sealed to prevent aeration. The samples were oven-dried (except samples containing wax) and weighed to determine the mass of the solid matrix of the sample and then placed in the vacuum dessicator. The vacuum dessicator was evacuated for 5 minutes, flushed with carbon dioxide for 5 minutes, and then evacuated again to remove all gas from the chamber. The process of evacuating and carbon dioxide flushing was repeated three times, and then the samples were saturated under 500 mm (19.7 in.) of mercury from the bottom up to allow entrapped soil gas to escape at sample surface during water imbibition.

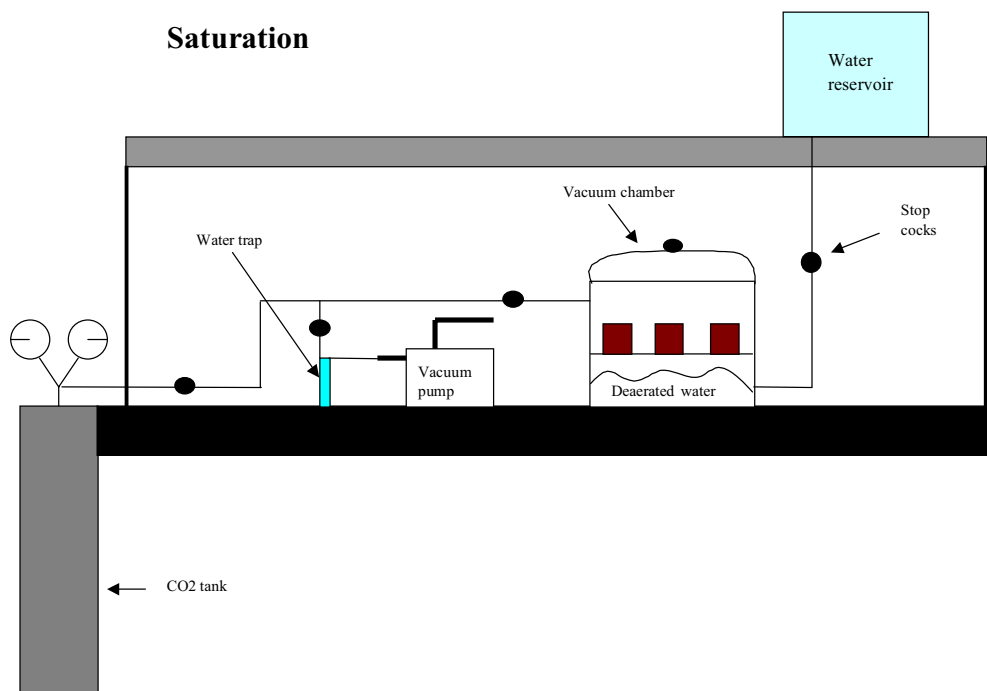


Figure C-2. Saturation chamber, vacuum pump, and carbon dioxide tank.

Once the water level reached midway up the sample length, the chamber was sealed, and the samples were left in the chamber for 24 hours under a vacuum to allow the sample to reach complete saturation. After 24 hours, the samples were removed from the chamber, and excess water around the edges of the samples was wiped off, taking care not to wick up any water from the sample pores. The sample then was weighed to determine the mass of the imbibed water. The volume of water was calculated from the sample water mass, assuming a density of 1 g/cm³. Porosity was determined by dividing the volume of the water in the sample (wet sample weight – dry sample weight) by the total sample volume.

C-4. DATA MANAGEMENT, ANALYSIS, AND INTERPRETATION

Data were recorded in a laboratory notebook (Logbook Number ER-108-2003), which includes procedures, raw data, and some calculations. The majority of the calculations were performed and recorded in an Excel spreadsheet, which can be provided upon request.

Figure C-3 shows the results of the porosity tests. Error bars represent the 95% confidence interval. The data are listed in Table C-2. Measured values of porosity for surface soil from the INL Site range from approximately 0.4 to 0.5 and are represented by the line shown in Figure C-3 at 0.45.

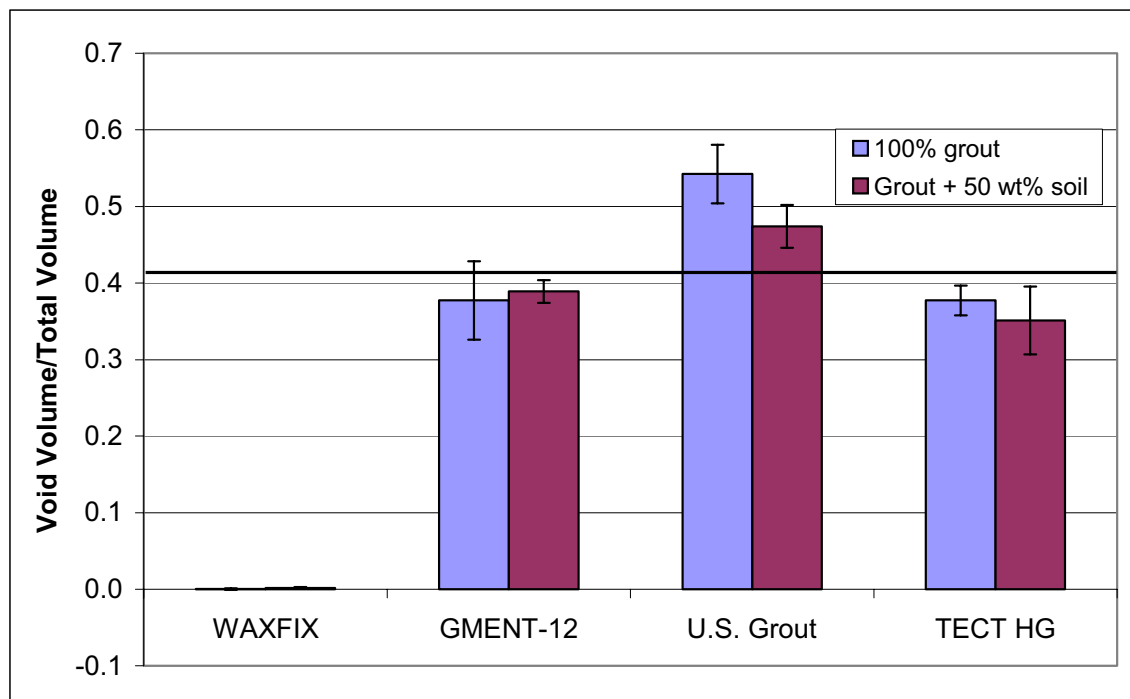


Figure C-3. Porosity of grouted waste forms.

Table C-2. Sample porosity for neat grout and soil from the Idaho National Laboratory Site.

Grout Type	Grouted Waste Form Tested	
	Neat Grout Porosity (mL/mL)	Grout and Soil Porosity (mL/mL)
WAXFIX		
A	0.0017	0.0003
B	0.0021	0.0001
C	0.0021	0.0006
Mean	0.0020	0.00035
Standard deviation	0.00021	0.00022
95% confidence interval	9.22E-04	9.05E-04

Table C-2. (continued).

Grout Type	Grouted Waste Form Tested	
	Neat Grout Porosity (mL/mL)	Grout and Soil Porosity (mL/mL)
GMENT-12		
A	0.3341	0.3801
B	0.3300	0.3888
C	0.4353	0.3799
D	0.4183	0.3749
E	0.4098	0.3992
F	0.3360	0.4119
Mean	0.3772	0.3808
Standard deviation	0.0488	0.0059
95% confidence interval	5.12E-02	1.48E-02
U.S. Grout		
A	0.5055	0.4817
B	0.5306	0.4861
C	0.5271	0.4707
D	0.5783	0.4370
E	0.5706	0.4949
F	—	0.5055
Mean	0.5424	0.4741
Standard deviation	0.0309	0.0225
95% confidence interval	3.83E-02	2.79E-02
TECT HG		
A	0.3924	0.1624
B	0.3898	0.3143
C	0.3960	0.3240
D	0.3559	0.3549
E	0.3834	0.3592
F	0.3517	—
Mean	0.3773	0.3308
Standard deviation	0.0185	0.0216
95% confidence interval	1.94E-02	4.40E-02

C-5. RESULTS AND CONCLUSIONS

The results show that neat WAXFIX samples had a significantly lower measured porosity relative to the other neat grout samples tested. All the grout-treated samples had a lower porosity than untreated soil from the INL Site, except the U.S. Grout-treated samples, which had a slightly higher porosity than the soil from the INL Site alone. Based on these test results, we conclude that all the grout evaluated (except the U.S. Grout) should reduce soil porosity when thoroughly mixed at the waste loading used in this test; however, WAXFIX performed much better at reducing the porosity of the soil from the INL Site than the cement-based grout tested and reduced the porosity even at the higher waste loading tested (70 wt% soil) with the WAXFIX.

C-6. REFERENCES

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Appendix D

Hydraulic Conductivity Tests—Part 1

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Appendix D

Hydraulic Conductivity Tests—Part 1

Hydraulic conductivity describes the ability of a porous medium to transmit fluids. The magnitude of hydraulic conductivity depends primarily on size distribution, roughness, tortuosity, shape, and degree of interconnection of the fluid-conducting pores. Because hydraulic conductivity is highly sensitive to texture and structure of the porous medium, mixing low-permeability grout materials into soil and soil-waste mixtures should significantly reduce the hydraulic conductivity of the waste forms, therefore, reducing mobility of radioactive and hazardous contaminants in the waste buried at the Subsurface Disposal Area.

Hydraulic conductivity testing will be used to evaluate potential application of in situ thermal desorption (ISTD) and in situ grouting to remediate transuranic pits and trenches, low-level waste pits and trenches and soil vaults, and ex situ grouting of Pad A salt waste. Comparison of test results will demonstrate relative efficiency in reducing hydraulic conductivity and potential for contaminant transport. The data also will be used to evaluate long-term physical stability of the grouted waste forms. The data will support modeling to estimate the release rate of contaminants from treated waste and comparison with the predicted release rate of untreated waste.

D-1. TEST OBJECTIVES, RATIONALE, AND DEVIATIONS FROM TEST PLAN

Two general laboratory methods are used for determining saturated hydraulic conductivity of porous materials: constant head (for porous media with high permeability) and falling head (for porous media with a low permeability). Column permeameters are used in both procedures. Because the samples for this study are expected to have low permeability, the falling-head method using a flexible wall permeameter was selected to measure hydraulic conductivity of mixtures of waste and grout and of neat grouts (ASTM 5084-00).

The flexible wall permeameter accurately measures flow through low-permeability porous media because flow along the edges of the sample holder is prevented by pressure applied along the sample holder flexible walls. However, the disadvantage of this method is that it is time intensive. To reduce the measurement time, ASTM 5084-00 was modified based on Dane and Topp (2002). Instead of using flexible wall permeameters, the modified method used Delrin plastic sleeves. Sample molds were prepared and then sealed into the plastic sleeves using epoxy to prevent preferential flow along the sample edges.

D-2. MATERIALS AND METHODS

Four in situ grouts and four surrogate types are being investigated for immobilization of subsurface contaminants at the Idaho National Laboratory (INL) Site. The grouts include GMENT-12, U.S. Grout, TECT HG, and WAXFIX. The surrogate types include nonradioactive organic sludge, nitrate salt sludge, uncontaminated soil from the INL Site, and ash from thermally treated organic sludge surrogate (see Appendix A for surrogates and recipes used in these tests). Loomis et al. (2003) previously tested hydraulic conductivity of the surrogate form mixtures for organic sludge, nitrate sludge, and soil from the INL Site and three of the neat grouts (GMENT-12, U.S. Grout, and TECT HG). Therefore, the waste forms bench-tested at the Science and Technology Complex for hydraulic conductivity include neat WAXFIX and WAXFIX-treated waste forms of organic sludge, nitrate sludge, and soil from the INL Site.

Hydraulic conductivity also was measured for thermally treated organic sludge mixtures using all the grouts under investigation.

D-2.1 Sample Preparation

Grout and waste mixtures were prepared according to predetermined maximum loading percentages to maintain a cohesive monolith and an adequate compressive strength of 250 psi (see Table D-1 for WAXFIX samples and Table D-2 for ISTD waste simulant). The WAXFIX mixtures of grout and waste and the neat WAXFIX samples were prepared by heating the materials and pouring them into plastic molds 25 mm in diameter and 45 mm high (see Figure D-1). The samples were cooled for a minimum of 48 hours. The Portland cement-based samples were cured for at least 30 days at 98 to 99% humidity in a constant humidity environmental chamber. Once the samples cured, they were removed from the molds, the edges were roughed, and the samples were sealed into plastic sample holders using J-B Weld (Sulphur Springs, Texas) epoxy.

Table D-1. Summary of hydraulic conductivity testing for in situ grouting of waste from transuranic pits and trenches.

Test Method	Measurement or Analytical Method	Waste Matrix	Analytes	Grout ^a	Waste in Grout (wt%)	Replicates	Total Samples
Cold Surrogates							
ASTM 5084-00 ^b	Falling-head method ^c	Organic sludge surrogate	N/A	WAXFIX	9	3	3
ASTM 5084-00 ^b	Falling-head method ^c	Nitrate salt sludge surrogate	N/A	WAXFIX	60	3	3
ASTM 5084-00 ^b	Falling-head method ^c	Soil from the INL Site	N/A	WAXFIX	70	3	3

a. WAXFIX will contain B-10.
b. 1990 version of test procedure is used for consistency with earlier testing.
c. Modified method (deviation from ASTM 5084-00) based on Dane and Topp (2002).

Table D-2. Summary of hydraulic conductivity testing for in situ grouting of ISTD-treated waste from transuranic pits and trenches.

Test Method	Measurement or Analytical Method	Waste Matrix	Analytes	Grout ^a	Waste in Grout (wt%)	Replicates	Total Samples
Cold Surrogates							
ASTM 5084-00 ^b	Falling-head method ^c	ISTD-treated organic sludge surrogate	Water	GMENT-12, U.S. Grout, TECT HG, WAXFIX	30 30 30 60	3	12

a. WAXFIX contains B-10.
b. 1990 version of test procedure is used for consistency with earlier testing.
c. Modified method (deviation from ASTM 5084-00) based on Dane and Topp (2002).



Figure D-1. Delrin plastic molds, 25-mm diameter by 45-mm height, used as sample holders for the waste forms.

D-2.2 Test Design and Procedures

In a falling-head permeameter, water is introduced to a saturated sample column by gravity drainage from a burette or standpipe, while the head on the downstream end remains constant (see Figure D-2). To prevent preferential flow along the edge of the sample holders using the falling-head method, the samples were epoxied into Delrin plastic sample rings measuring 25 mm in diameter by 45 mm high (see Figure D-1).

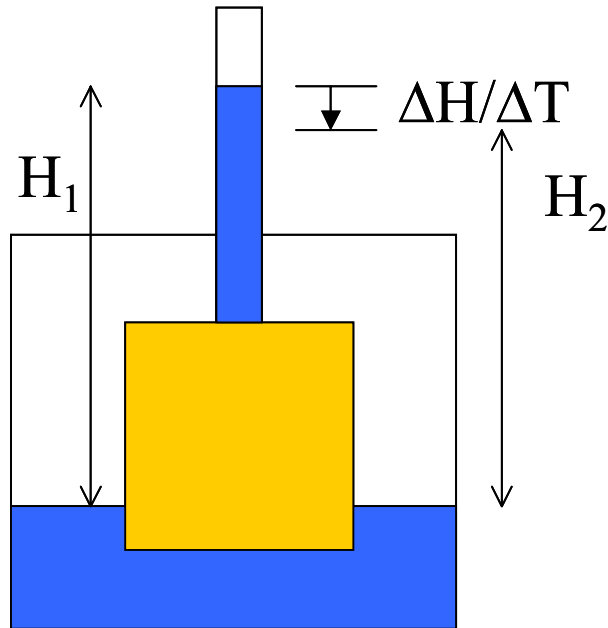


Figure D-2. Falling-head method for measuring hydraulic conductivity of low-permeability porous media.

To test for preferential flow along the edges of the rings, the samples were pretested using red dye before starting the conductivity tests. The samples were then saturated under a vacuum (see Appendix C). A plastic cap with a water-saturated, 1/2-bar, ceramic-disk insert was placed on top of the sample, sealing with an O-ring to the sample ring. The ceramic disk was used to uniformly disburse the water across the top of the sample during infiltration. Permeability of the disk was greater than the sample material and thus did not influence measurement results.

A 10-mm-diameter burette was attached to the top of the sample cap. All air was removed from the porous-ceramic disk, the cap, and the tubing connecting the cap to the burette. The sample was placed in a beaker of water, and the burette was filled with water to the 0-ml mark. The initial distance from the water level in the burette and the water in the beaker was determined for each sample (ΔH). The permeameters were placed in a humidity chamber to minimize evaporation during testing. A blank permeameter (no waste form) was set up to test daily evaporation rates inside the humidity chamber.

Figure D-3 shows the actual experimental setup and instrumentation used for falling-head tests of all the grouts and grout-waste mixtures in this study. The water level in the burette was monitored daily over 1 week, and changes in hydraulic head were recorded. Data representing steady state conditions (i.e., constant flux and hydraulic gradient) were used to calculate hydraulic conductivity.

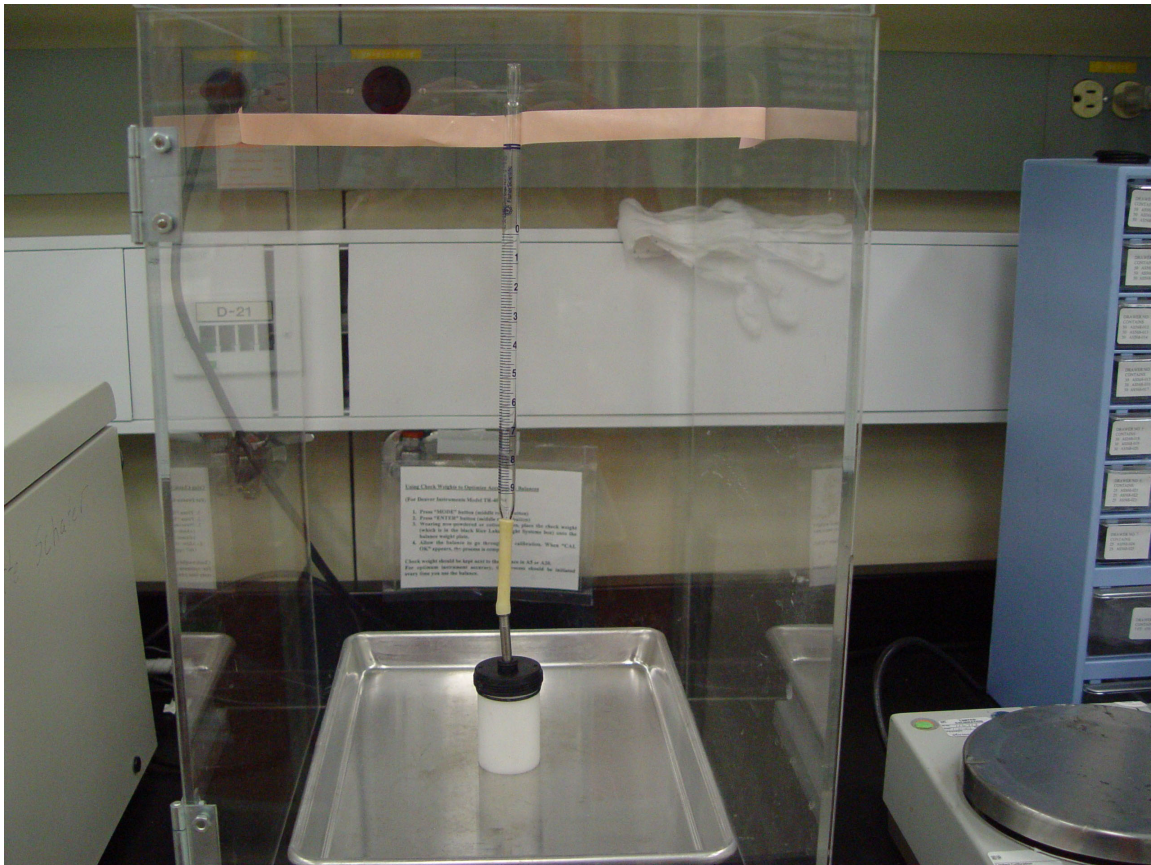


Figure D-3. Hydraulic conductivity measurement in 98 to 99% humidity controlled environmental chamber.

D-2.3 Data Management, Analysis, and Interpretation

Data were recorded in a laboratory notebook (Logbook Number ER-108-2003), which includes procedures, raw data, and some calculations. The majority of the calculations were performed and recorded in an Excel spreadsheet, which can be provided upon request. The hydraulic conductivity of a porous material is qualitatively defined as the ability of a saturated porous medium to transmit fluids. The physical relationship often used to describe fluid flow through porous materials is Darcy's law (Dane and Topp 2002). The saturated hydraulic conductivity of a porous medium can be calculated from direct measurements of flux, hydraulic head, and column dimensions.

Modification of Darcy's Law is often used to calculate hydraulic conductivity using the falling-head method (Stephens 1995) as shown in Equation (D-1):

$$K = \frac{aL}{At} \ln \left(\frac{H_1}{H_2} \right) \quad (D-1)$$

where

- A = sample cross-sectional area [L²]
- L = sample length [L]
- a = burette's cross-sectional area [L²]
- t = time between measurements
- H₁ and H₂ = water levels in the burette at t₁ and t₂, respectively.

Data were quality checked before being entered into an Excel spreadsheet, and then were checked again after entering.

D-3. RESULTS AND CONCLUSIONS

In many cases, the data for the first 24 hours show a relatively higher flux of water followed by a steady state flux for the remainder of the 7-day test. For this reason, the systems were allowed 24 hours to reach steady state before the data were used to calculate the hydraulic conductivity. The initial variability in flux was most likely from air present in the cap that was placed over the sample, or small pockets of air that were not filled during saturation of the sample. Data collected during the first 24 hours were not included in the calculations. Table D-3 shows the mean, standard deviation, and 95% confidence interval for the grout and waste types tested.

Table D-3. Hydraulic conductivity values for the grouts and surrogates.

Sample	Grout Product			
	WAXFIX	GMENT-12	TECT HG	U.S. Grout
Neat Grout				
A	4.16E-10	8.50E-09	9.80E-09	1.70E-08
B	<detection	6.10E-09	1.70E-09	1.90E-08
C	<detection	—	—	—
Mean	1.39E-10	7.30E-09	5.75E-09	1.80E-08
Standard deviation	2.40E-10	1.70E-09	5.73E-09	1.41E-09
95% confidence interval	+/-1.10E-10	+/-4.22E-09	+/-4.18E-09	+/-1.03E-09
Soil from the Idaho National Laboratory Site				
	70 wt%	50 wt%	50 wt%	50 wt%
A	7.24E-09	6.00E-09	2.00E-08	3.00E-09
B	4.04E-09	1.00E-08	8.00E-09	2.00E-08
C	1.09E-08	—	—	—
Mean	7.39E-09	8.00E-09	1.40E-08	1.15E-08
Standard deviation	3.43E-09	2.83E-09	8.49E-09	1.20E-08
95% confidence interval	+/-1.57E-09	+/-2.06E-09	+/-6.19E-09	+/-8.77E-09
Organic sludge				
	9 wt%	9 wt%	9 wt%	9 wt%
A	<detection	2.00E-09	5.00E-09	1.00E-08
B	<detection	4.00E-09	1.00E-09	2.00E-08
C	<detection	—	—	—
Mean	<detection	3.00E-09	3.00E-09	1.50E-08
Standard deviation	<detection	1.41E-09	2.83E-09	7.07E-09
95% confidence interval	<detection	+/-1.03E-09	+/-2.06E-09	+/-5.16E-09
Nitrate salt sludge				
	60 wt%	12 wt%	12 wt%	12 wt%
A	1.14E-08	5.00E-07	6.00E-09	7.00E-09
B	1.53E-08	7.00E-08	2.00E-08	2.00E-08
C	9.24E-09	—	—	—
Mean	1.20E-08	2.85E-07	1.30E-08	1.35E-08
Standard deviation	3.08E-09	3.04E-07	9.90E-09	9.19E-09
95% confidence interval	+/-1.45E-09	+/-2.22E-07	+/-7.22E-09	+/-6.70E-09
ISTD-treated organic sludge				
	60 wt%	30 wt%	30 wt%	30 wt%
A	<detection	2.75E-07	8.04E-09	1.41E-08
B	<detection	1.36E-08	2.00E-08	1.06E-08
C	4.07E-09	1.44E-08	1.32E-08	9.33E-09
Mean	1.36E-09	1.40E-08	1.38E-08	1.13E-08
Standard deviation	2.35E-09	5.96E-10	5.98E-09	2.44E-09
95% confidence interval	+/-1.16E-09	+/-2.99E-10	+/-2.54E-09	+/-1.15E-09

Test results show that neat WAXFIX and WAXFIX surrogate mixtures all had hydraulic conductivities lower than 10^{-7} cm/second (see Table D-3). The samples shown in Figure D-4 include neat WAXFIX, a 30-70 ratio of WAXFIX to soil from the INL Site, a 90-10 ratio of WAXFIX to organic sludge, and a 40-60 ratio of WAXFIX to nitrate salt sludge. (WAXFIX was tested at 70 wt% soil from the INL Site, 10 wt% organic sludge, and 60 wt% nitrate salts.) The WAXFIX/nitrate salt sludge mixture waste forms had the highest hydraulic conductivity of the four WAXFIX mixtures tested (see Figure D-4). This may have been due to dissolution of salts during water saturation. Comparison between soil from the INL Site and soil from the INL Site mixed with WAXFIX shows that uniform mixing of WAXFIX with the soil lowered the hydraulic conductivity of soil from the INL Site by three orders of magnitude (see Figure D-4).

For comparison purposes, Figure D-4 also includes the results of tests performed by Loomis et al. (2003) for the Portland cement-based grouts (GMENT-12, TECT HG, and U.S. Grout) and the grout-treated mixtures. The graph shows that the hydraulic conductivity of the WAXFIX neat grout and WAXFIX/organic sludge was significantly lower than the neat cement-base grouts and the other grout-treated mixtures. For the WAXFIX/organic sludge surrogate, no measurable flow was observed. This is likely because of the hydrophobic natures of both the WAXFIX and the organic sludge surrogate and because the organic sludge and WAXFIX were more compatible mixtures than the cementitious grouts and the organic sludge surrogate. All the grout-treated mixtures had a hydraulic conductivity less than 10^{-7} cm/second with exception to the GMENT-12/nitrate salt mixture. From the results of the hydraulic conductivity tests, GMENT-12 appears to not react well with the nitrate salts compared to the other grouts.

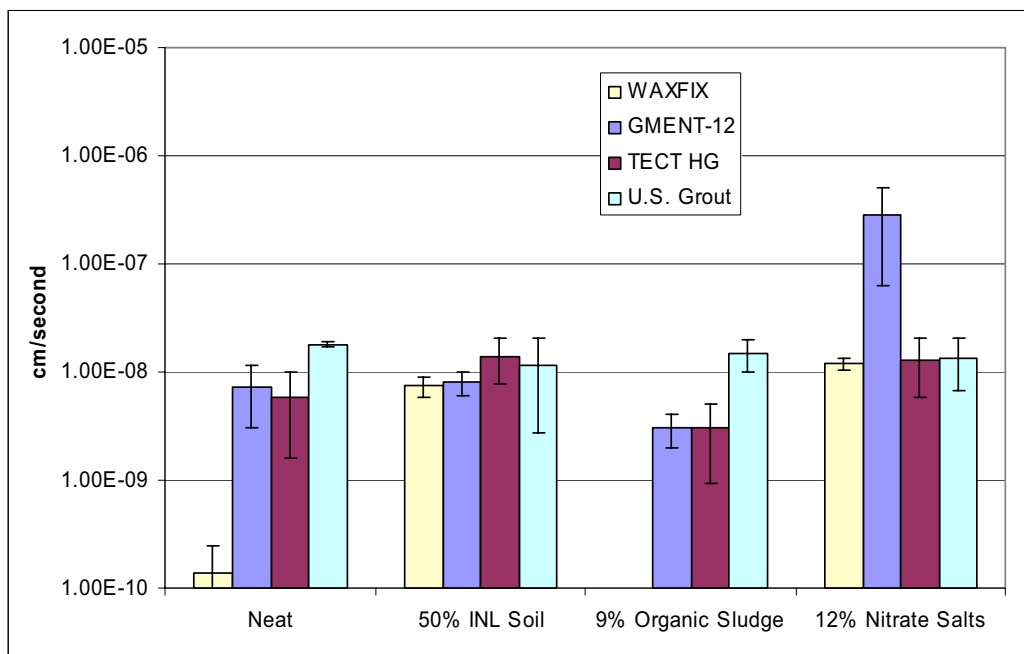


Figure D-4. Results of hydraulic conductivity testing of grouts and grout-tested surrogate, representing waste in Idaho National Laboratory Site transuranic pits and trenches.

Comparisons between the four grouts tested with thermally treated organic sludge show that all four had conductivity measurements lower than 10^{-7} cm/second. However, WAXFIX had the lowest conductivity of the four grouts when tested neat and when mixed with the thermally treated sludge (see Figure D-5). The hydraulic conductivity of all of the grouts was higher in the neat grout than when

mixed with the thermally treated organic sludge, except for U.S. Grout, where the neat grout had a higher hydraulic conductivity than the same grout mixed with ISTD-treated organic sludge. When you compare this with the porosity data in Appendix C, the data show that the porosity of the neat U.S. Grout is higher than the porosity of the soil and higher than the porosity of the other neat grouts. This coincides with the data here, where the hydraulic conductivity of the neat U.S. Grout is higher than the other grouts as well. Overall, the grout-treated mixtures for the thermally treated organic sludge were within an order of magnitude difference, suggesting all three grouts were comparable in reducing hydraulic conductivity of the surrogate (see Table D-3).

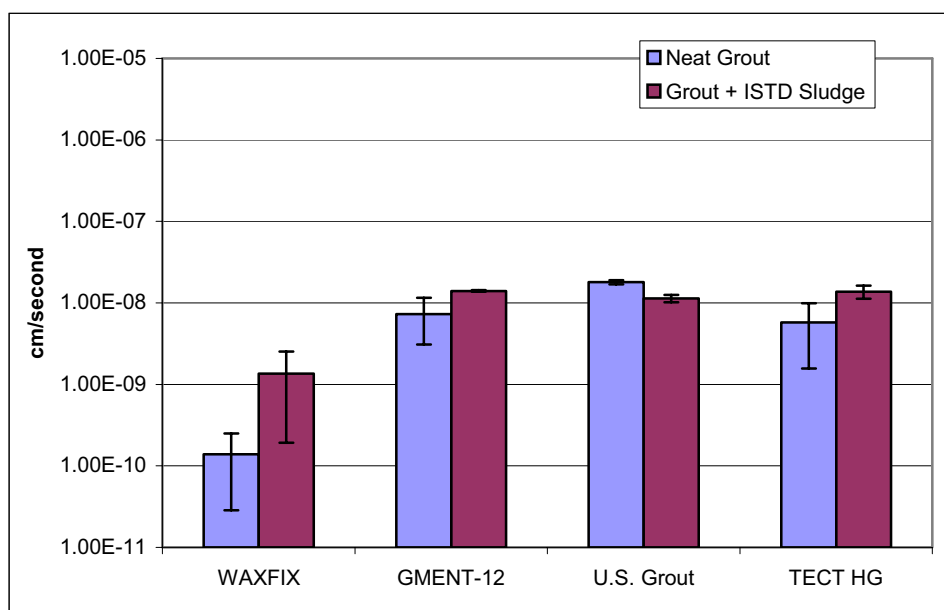


Figure D-5. Hydraulic conductivity of grout mixtures of ISTD-treated organic sludge.

In conclusion, the results from the hydraulic conductivity tests showed that although all the grouts tested significantly reduced the hydraulic conductivity of the surrogate tested (with exception to the GMENT-12/nitrate salt mixture), WAXFIX performed the best overall. Of the WAXFIX mixtures tested, the WAXFIX/nitrate salt mixture had the highest hydraulic conductivity, while the WAXFIX/organic sludge mixture had the lowest hydraulic conductivity.

D-4. REFERENCES

- ASTM 5084-00, 2001, "Standard Test Method for Measurement of Hydraulic Conductivity of Saturated Porous Materials Using a Flexible Wall Permeameter," ASTM International.
- Dane, J. H. and G. C. Topp, eds., 2002, *Methods of Soil Analysis Part 4 Physical Methods*, Madison: Soil Science Society of America, Inc.
- Loomis, Guy G., James J. Jessmore, Jerry R. Weidner, Christopher M. Miller, and Allen L. Sehn, 2003, *Final Results Report, In Situ Grouting Technology for Application in Buried Transuranic Waste Sites*, INEEL/EXT-02-00233, Rev. 1, Idaho National Engineering and Environmental Laboratory.

Appendix E

Boron Retention Tests

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Appendix E

Boron Retention Tests

One concern with using WAXFIX as a grout in the Subsurface Disposal Area (SDA) was the potential for the main ingredient of WAXFIX, paraffin, to act as a moderator when combined with radionuclides and raise the potential for a criticality occurring in the subsurface. The boron retention tests evaluated the potential for adding boron, a moderator poison, to the WAXFIX to eliminate the concerns of the grout acting as a moderator.

E-1. TEST OBJECTIVES, RATIONALE, AND DEVIATIONS FROM TEST PLAN

The main objective of this test was to determine the extent to which boron is retained in the WAXFIX grout as it flows through soil and cools. One of the issues is that the moderating properties of WAXFIX grout could increase the potential for a criticality in the transuranic pits and trenches. The solution to this potential issue is the addition of B-10 (a poison for nuclear reactions) at 1 g/L in the molten wax and ensuring that the distribution remains at that concentration during the cooling process.

Boron retention and distribution in WAXFIX were tested using cold surrogates. The concentration of B-10 is based on criticality calculations for emplacement in and around a postulated critical mass of Pu-239 (Farnsworth et al. 1999). The B-10 is essentially inert in the wax and is expected to be functional as long as the WAXFIX remains intact. The laboratory tests included slow (5 days) cooling of WAXFIX with B-10 alone and potential filtering of B-10 from WAXFIX that has migrated into a column of soil. The purpose of the soil test was to assess the potential for B-10 to be filtered from the WAXFIX as it migrates from the original placement site during cooling.

In August of 2003, *Criticality Safety Study of the Subsurface Disposal Area for Operable Unit 7-13/14* was published (Sentieri and Taylor 2003), which stated that the presence of paraffin grout within the SDA would not lead to the formation of a critical mass. At this point, testing had begun using density changes in the WAXFIX to determine loss of boron. The next step was to perform a chemical analysis to validate the method of using density change to determine boron loss. However, because of the finding from Sentieri and Taylor (2003), it was no longer necessary to complete the boron retention testing.

E-2. EXPERIMENTAL DESIGN AND PROCEDURES

For this test, two experiments were conducted. The first was to pour molten WAXFIX into a column (see Figure E-1), allow the WAXFIX to cool slowly over a 5-day period, and once cooled, measure the amount of separation that occurred by the boron in the WAXFIX.

The second test consisted of pouring molten WAXFIX through a column of soil (see Figure E-2) and determining if boron flowed through the soil with the WAXFIX or if it was filtered out by the soil.



Figure E-1. Boron separation test in molten WAXFIX.



Figure E-2. Boron separation test using a soil column.

E-3. EQUIPMENT AND MATERIALS

Boronated WAXFIX was purchased from Carter Technologies. The WAXFIX was heated in an oven at 120°C (248°F). The molten boronated WAXFIX was poured into a 2.5-cm (1-in.)-diameter column. The temperature in the oven was maintained at 120°C (248°F) for 1 day. Each day for 5 days, the temperature was reduced until the 5th day, when the oven was turned off and allowed to reach ambient temperature ~20°C (~68°F).

To determine the separation of boron from WAXFIX as it flowed through a column of soil, a 2.5-cm (1-in.)-diameter column of soil was heated to 120°C (248°F). Molten WAXFIX at 120°C (248°F) was allowed to flow through the soil column. Boron loss was determined in the WAXFIX after passing through the heated soil.

E-4. DATA MANAGEMENT, ANALYSIS, AND INTERPRETATION

Loss of boron from the WAXFIX was measured by changes in density of the wax. As the boron settled from the WAXFIX or was filtered out by the soil, the density of the WAXFIX decreased. That decrease was used to directly determine the amount boron lost from the sample. Additionally, more quantitative measurements were planned, but the work was not completed since the concern for paraffin acting as a moderator was eliminated.

E-5. RESULTS AND CONCLUSIONS

Sentieri and Taylor (2003) concluded that a criticality formation caused by the presence of paraffin grout within the SDA would not be credible. Based on this, any further testing for boron was canceled. The only results that were measured were the density change results. The project was in the process of performing analytical measurements to verify the results from changes in density. As a result, only the density data are presented in this section.

The results of the boron separation from molten WAXFIX showed that, over a 5-day cooling period, there is a small amount of separation that occurs. Figure E-3 shows that, over a 47-cm (18.5-in.) column, less than 1 wt% of the boron had settled.

Loss of Boron in Molten WAXFIX while Slowly Cooling

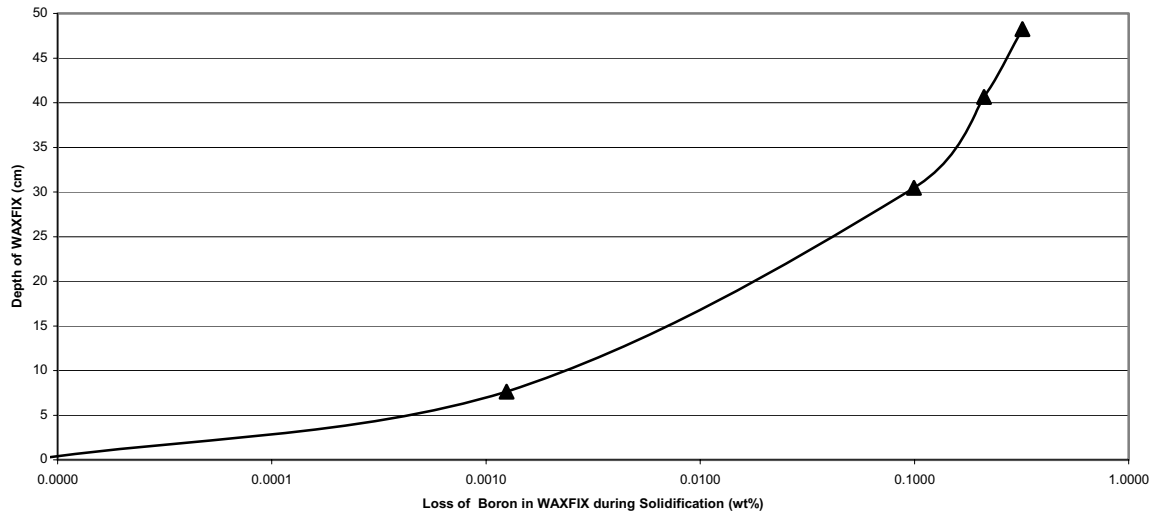


Figure E-3. Weight percent loss of boron during cooling.

A significant amount of separation occurred as the WAXFIX flowed through the soil. Figure E-4 shows the amount of boron lost as a function of the amount of soil the WAXFIX passed through. Figure E-4 shows that as little as 7.6 cm (3 in.) of soil will bring the boron below the 1-g/L recommended concentration. By the time the WAXFIX has traveled through the 47-cm (18.5-in.) column, almost all of the boron has been filtered out.

Loss of Boron in WAXFIX Flowing Molten through Heated SDA Soil

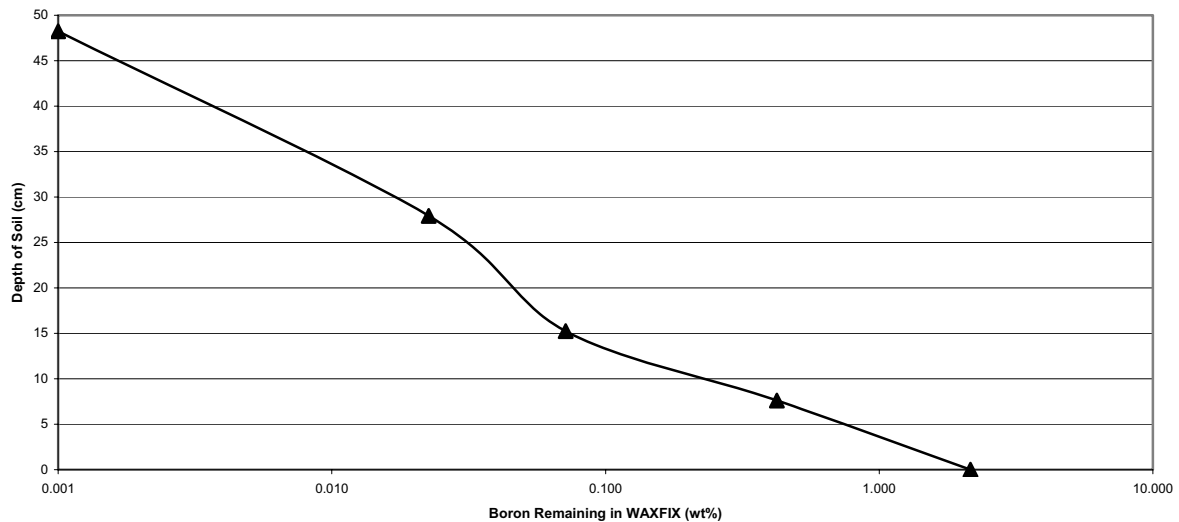


Figure E-4. Loss of boron in molten WAXFIX through heated soil from the Idaho National Engineering and Environmental Laboratory Site.

E-6. REFERENCES

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Appendix F

U.S. Department of Transportation Oxidizer Tests

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Appendix F

U.S. Department of Transportation Oxidizer Tests

The U.S. Department of Transportation (DOT) oxidizer tests (see Section 4.3.3.2 of Yancey et al. [2003]) evaluated whether WAXFIX increases the potential for oxidation of nitrate salts (the primary ingredient of the Pad A nitrate salts). Determining the enhanced or decreased flammability supports the evaluation of WAXFIX for use as grout with the nitrate salt waste.

F-1. TEST OBJECTIVES, RATIONALE, AND DEVIATIONS FROM THE TEST PLAN

Because the potential for a rapid reaction is present if nitrates are involved in an accidental fire during storage or shipping, the objective of these tests was to determine whether sodium nitrate encapsulated in paraffin (WAXFIX) would be classified as a DOT oxidizer. The tests were “. . . designed to measure the potential for a solid substance to increase the burning intensity of a combustible substance when the two are thoroughly mixed. . .” (Milian et al. 1997).

Previous data for DOT oxidizer tests on paraffin were located. Therefore, the laboratory test was not completed. A summary of the previous data is provided in this section.

F-1.1 Experimental Design and Procedures

The recommended test procedure for quantifying hazards associated with solid oxidizing materials is identified in “Shippers—General Requirements for Shipments and Packaging” (49 CFR 173). The definition of an oxidizer is found in “Class 5, Division 5.1 Definition and Assignment of Packing Groups” (49 CFR 173.127). The procedure is found in Appendix F of latter regulation. Test materials were prepared in the following compositions:

- Refined paraffin wax cut and sieved to a particle size less than 2 mm and mixed with sodium nitrate in a mass ratio of 1 to 1
- Sodium nitrate salt encapsulated in refined paraffin wax in a mass ratio of 1 to 1
- Sodium nitrate salt encapsulated in refined paraffin wax in a mass ratio of 1 to 1, but cut to a sieve-mesh size less than 9.5 mm following solidification
- 100% sodium nitrate salt.

A combustible material, wood sawdust, was added to each test material in mass ratios of 1 to 1 and 4 to 1. All tests were conducted according to the previously indicated DOT test procedures (Milian et al. 1997).

F-1.2 Equipment and Materials

See Section F2.

F-2. DATA MANAGEMENT, ANALYSIS, AND INTERPRETATION

Published data were used (Milian et al. 1997).

F-2.1 Results and Conclusions

Results in Table F-1 (Milian et al. 1997) indicate the nitrate salt encapsulated in paraffin wax, either as one large piece or sieved to less than 9.5 mm, burned significantly slower and less violently than the sodium nitrate without wax. Similarly, the nitrate salt mixed with chopped paraffin wax resulted in much slower burn times. Based on these results, sodium nitrate solidified in solid paraffin wax or mixed with paraffin wax is not classified as an oxidizer based on the recommended DOT tests.

Table F-1. Results from the U.S. Department of Transportation oxidizer tests.

Combustion Time for Test Materials		
Material Composition (wt%)	1 to 1 Mass Ratio Test Mixture	4 to 1 Mass Ratio Test Mixture
	Burn Time (seconds)	Burn Time (seconds)
50% chopped wax (less than 2 mm)/50% sodium nitrate	161 ^a	188 ^a
Encapsulation: 50% Wax/50% sodium nitrate	131 ^a	819 ^a
Encapsulation: 50% Wax/50% sodium nitrate and sieve to less than 9.5 mm	628 ^b	525 ^b
100% sodium nitrate	37 ^b	25 ^b
a. Mean of two replicates.		
b. Based on one replicate.		

F-3. REFERENCES

- 49 CFR 173, 2003, "Shippers—General Requirements for Shipments and Packagings," *Code of Federal Regulations*, Office of the Federal Register.
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Appendix G

Hydrogen-Generation Tests

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Appendix G

Hydrogen-Generation Tests

Yancey et al. (2003) identified generation hydrogen from radiolysis of paraffin in WAXFIX as a potential issue for the long-term effectiveness of WAXFIX.

G-1. TEST OBJECTIVES, RATIONALE, AND DEVIATIONS FROM TEST PLAN

The main objective of the hydrogen-generation test was to obtain data about the alpha component of hydrogen production from the WAXFIX material. The predominant alpha materials from Radioactive Waste Management Complex—U-238, U-235, and Pu-239—do not have a high enough alpha emission rate for a short-term test to generate enough hydrogen to measure. Two less common radioisotopes found at the Radioactive Waste Management Complex do have a satisfactory alpha decay rate: Pu-238 and U-233. Uranium-233 was selected because it would have been difficult to get permission to work with plutonium within a reasonable timeframe.

Uranium-233 in a solid dioxide form was available at the Idaho National Laboratory Site. An estimate was calculated of the alpha emissions from solid particulates containing the alpha-generating nuclides. This spreadsheet was used in determining the nuclides that could be used and the desired mass and particulate size necessary to generate a measurable amount of hydrogen during the allotted test time.

G-2. EXPERIMENTAL DESIGN AND MATERIALS

Uranium-233 in the form of uranium dioxide was used in the tests. The uranium oxide contained other uranium isotopes plus the decay daughters. See Table G-1 for the uranium isotopes as shipped (in accordance with shipping papers) and decayed to the start of testing on March 12, 2004.

Table G-1. Activity of uranium oxide materials initially and at start of testing.

Isotope	Original Material		Decayed to Start of Testing (3/13/04)
	Total Uranium (wt%)	Uranium Oxide (Ci/g)	Uranium Oxide (Ci/g)
U-232	2E-04	9.67E-05	7.88E-05
U-233	98.22	8.32E-03	8.32E-3
U-234	1.111	6.1E-05	6.1E-5
U-235	0.39	7.4E-10	7.4E-10
U-236	0.002	1.14E-09	1.14E-9
U-238	0.624	1.85E-09	1.85E-9

The total mean energy of emissions, separated by type (α , β , and γ), for the uranium oxide and the primary isotopes and decay daughters contributing to the α term are presented in Table G-2.

Table G-2. Total energy emission rate and primary α energy rate contributors for uranium oxide.

	Uranium Oxide (MeV/g/second)	Total Sum (%)
Total α	1.645E+09	98.2
Total β	8.592E+06	0.51
Total γ	2.152E+07	1.28
U-233 α	1.504E+09	89.8
U-234 α	1.074E+07	0.64
U-233 Daughters α	1.975E+07	1.18
U-232 + Daughters α	1.113E+08	6.6

The U-233 α accounts for about 90% of the energy emission from the uranium oxide sample. About 7% comes from U-232 α and daughter decays. Nearly all the balance of the energy is from U-233 daughter and U-234 decay. The high proportion for U-232 is because of its short half-life of 68.9 years, which results in it being near equilibrium with its short-lived daughters.

The test was designed to produce enough hydrogen gas from alpha radiolysis for it to be detectable within 24 hours with a detection limit of 10 ppm. The experiment had four separate alpha activities of diminishing amounts so that the smallest was less than 4% of the largest. This spacing was necessary to produce hydrogen so that any effects that might be caused by a variable alpha dose to the WAXFIX would be evidenced. Spacing of the amount of uranium oxide also was needed because variation in the uranium oxide particle size distribution, which was unknown, would cause variation in the hydrogen production rate caused by internal absorption of α energy by the uranium oxide particles. Larger particles may release only a fraction or none of the total α decay energy. The use of four generators also allowed statistical information to be collected.

The maximum dose was fixed by the amount of the U-233 material that would be allowed in an unshielded area (\cong 0.4 g [0.01 oz]). This amount, with the detection limit, fixed the volume of the containers to no more than 250 mL (15.3 in.³). To ensure the hydrogen concentration in the reactors was above the detection limit of the gas chromatograph, the volume of the reactors was decreased to 60 mL (3.7 in.³).

A sketch of the alpha-WAXFIX hydrogen generator is shown in Figure G-1. Glass vials contained the WAXFIX uranium dioxide mixtures. Interaction of the alpha with the glass will not produce hydrogen but is likely to deflect the alpha back into the WAXFIX. The vials were large enough (diameter and length) so that few alpha particles free of the uranium oxide would escape before interacting with the WAXFIX. Escape of alpha from the WAXFIX on the inside surface is most likely to result in collision of the alpha with the WAXFIX on the opposite side.

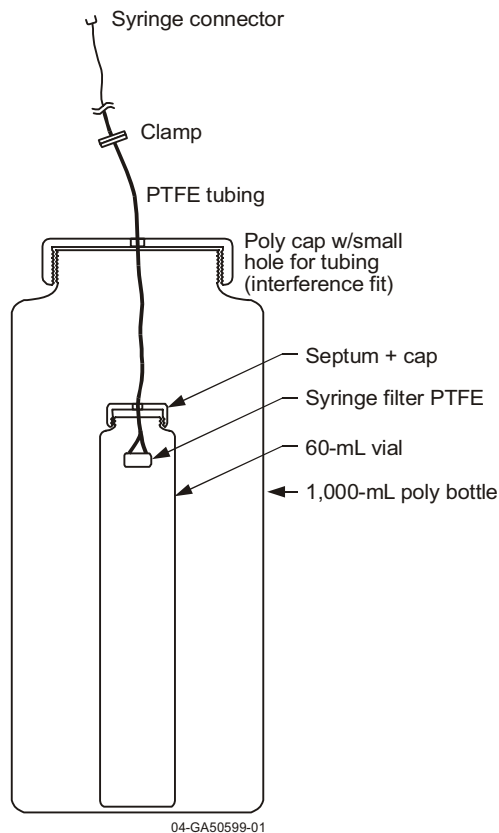


Figure G-1. Diagram of apparatus.

The plan was to mix thoroughly the uranium oxide particles with the wax in a thin layer on the inside wall of the 60-mL (3.7-in.³) vials. An aliquot of 3 g (0.1 oz) of WAXFIX was used on the inside of each of the six vials: four generators containing uranium oxide and two blank vials containing only WAXFIX. The calculated thickness of the mixture applied to the entire cylindrical surface would be 1.7 mm (0.07 in.). A thin film allows for easier escape of the hydrogen from the WAXFIX. As applied, the wax and uranium-oxide mixture was not as evenly distributed as during trial testing using zirconium oxide. The uneven distribution was caused by the large particle size and the resistance of the uranium oxide to “wet” easily with the wax. Finer uranium oxide in the sample distributed better. In general, the wax distributed evenly on the vial walls.

The amount of WAXFIX in each generator contains about eight million times the total hydrogen that could be produced by the total alpha emitted in 10 years for the highest dose generator. This should more than suffice for 2 months of testing.

Each of the four uranium-WAXFIX generator systems and the two blank systems were handled in the same manner. After mixing the sample and wax, each generator system was evacuated in turn—using a vacuum pump—and purged with purified argon three times to remove air. This was done to mitigate the hydrogen from combining with oxygen, to allow detection of air in leakage into the vials during sampling, and to ensure sample integrity during analysis, and served as the starting point for the test (time zero). After the purging of each vial was complete, the flex tubing was clamped, and the set (purge) syringe designated for that generator was attached. The tubing was unclamped, and the 2 mL (0.12 in.³) of argon in the syringe pushed into the system to supply a slight overpressure relative to atmospheric pressure. This pressure was applied to inhibit atmospheric gases leaking in and to test for possible leaks.

After 30 minutes, the indicated volume in the syringe resulting from pressure “push back” was recorded. This value was consistent until near the end of the experiment at 0.2 mL (0.012 in.³). Near the end, the value decreased to about 0.1 mL (0.006 in.³). This volume measurement is not a true indicator of pressure because of friction from the syringe plunger.

At chosen intervals, samples were drawn from the generators in the following sequence:

1. The purge syringe was drawn back to 2 mL (0.12 in.³) to purge the sample line and filters.
2. A clamp was placed on the flex tubing.
3. The purge syringe was removed and the sample syringe attached. The clamp was removed, and the plunger of the sample syringe was drawn back to 1 mL (0.06 in.³).
4. Meanwhile, the purge syringe was evacuated and refilled with 3 mL (0.18 in.³) of argon. Purified nitrogen was used later.
5. When the purge syringe was ready, the valve on the sample syringe was closed, and the clamp was put on the flexible tubing.
6. The purge syringe was reattached, the clamp was removed, and the 3 mL (0.18 in.³) of purge replacement gas was pushed into the system. For purposes of measurement and system enclosure, the purge syringe remained in place until the next sample was drawn.

A sample syringe from each of the vials (four test and two control) was submitted that day or the next for analysis by gas chromatograph.

G-3. EQUIPMENT AND MATERIALS

The preparation procedure for the hydrogen is detailed in the job safety analysis. A sketch of the apparatus is shown in Figure G-1. A description of the setup is as follows: Septum fitted caps closed interior volumes of the vials. A small, 21-gauge, tetrafluoroethylene (TFE) (i.e., Teflon) sampling tube was passed through the septum. On the inside of the vial cap, a TFE syringe filter was attached to the tubing using an interference fit. The filter was used to keep nongaseous contaminants (particularly the radioactive ones) out of the gas sample. An enclosure bottle (1,000 mL [61 in.³]) for each vial was provided for protection of the hydrogen generators and for radiological contamination control. The TFE sampling tube ran through the lid of the enclosure bottle and out of the fume hood to the sampling table. At the end of each TFE tube, a small-diameter, flexible tygon tubing section was attached by an interference fit. This section of flexible tubing was required to isolate the system during the sampling procedure. To the sampling end of the flexible tubing, a 21-gauge hypodermic needle was inserted (interference fit). A syringe filter was attached to the Lure-Lok needle that also had a Lure-Lok adapter. This adapter could accommodate the 3-mL (0.18-in.³) purge syringe and the adapted 1-mL (0.06-in.³) Hamilton gas sample syringe. A replica of the sample tubing, filters, and needle was tested for volume. The total volume was 0.57 ± 0.1 mL (0.035 ± 0.006 in.³). Each of the six systems was checked for leaks by pressurization and by pulling a vacuum before final assembly with the WAXFIX and uranium-oxide mixture. This was accomplished using a 60-mL (3.7-in.³) syringe.

Pictures of the vials containing WAXFIX and uranium oxide are shown as Figures G-2 and G-3.

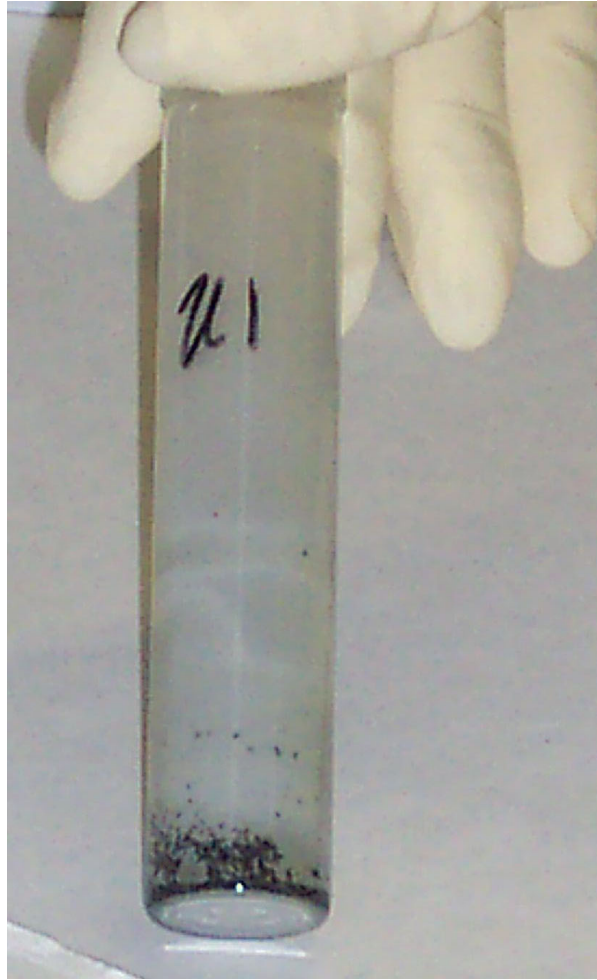


Figure G-2. U1 generator.

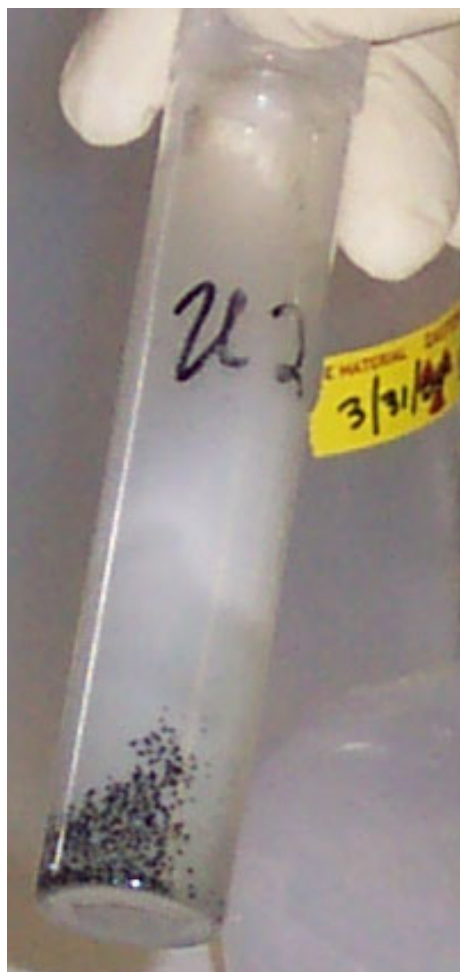


Figure G-3. U2 generator.

All the uranium oxide used came from a 10-g (0.35-oz) U-233 shipment that came from Oakridge National Laboratory in 1983. It is assumed to be of uniform composition, but it is not of uniform size distribution. Samples were obtained for analyses, but time restraints did not allow for a complete analysis.

G-4. DATA MANAGEMENT, ANALYSIS, AND INTERPRETATION

All gas samples drawn from the generators were run through a gas chromatograph—Hewlett-Packard Model 5890 Series II—with a 25-m by 0.53-mm (82-ft by 0.02-in.) ID molesieve column. This instrument was calibrated using an hydrogen standard diluted with purified nitrogen. A single point calibration was used for the first and second sample pull. All subsequent samples were determined from a three-point calibration. A calibration check was made before running each sample group.

As was mentioned previously, four different generators (and two blanks) were prepared with diminishing masses of uranium oxide. Table G-3 provides the masses and calculated total alpha energy emission rates.

Table G-3. Generator mass and total alpha energy emission rate.

Generator	Mass Uranium Oxide	Total Alpha Energy Emission Rate—MeV/second
U1	0.163 ± 0.002	2.68E+08
U2	0.078 ± 0.002	1.28E+08
U3	0.016 ± 0.002	2.63E+07
U4	0.006 ± 0.001	9.87E+06
B1	0	0
B2	0	0

The gas sampling occurred mostly at weekly intervals. Shorter intervals were used at the beginning. The sample schedule showing the raw sample results is included in Table G-4. The calculated hydrogen production rates, obtained from the data of Table G-4, are represented by data points on Figure G-4.

Table G-4. Unadjusted raw data from hydrogen generators.

Generator	Grams UO ₂	1st Sample H ₂ Concentration ppm	2nd Sample H ₂ Concentration ppm	3rd Sample H ₂ Concentration ppm	4th Sample H ₂ Concentration ppm	5th Sample H ₂ Concentration ppm	5th-a Sample H ₂ Concentration ppm	6th Sample H ₂ Concentration ppm	7th Sample H ₂ Concentration ppm	8th Sample H ₂ Concentration ppm
Date of Sample	4/5/04	4/6/04	4/8/04*	4/12/04	4/19/04	4/26/04	4/28/04	5/3/04	5/10/04*	5/12/04
U1	0.1625	25.9		138.4	592.3	609.2	Not sampled	736		762.4
U2	0.078	ND		16.5	No Ar	No Ar	88.7	97.8		108.2
U3	0.016	ND		6.4	48.9	52.3	Not sampled	62.4		53.6
U4	0.006	ND		Trace	19.9	27.2	Not sampled	16.6		19.7
B1	0	ND		ND	ND	ND	Not sampled	U4b1 16		Not sampled
B2	0	ND		ND	ND	ND	U2b2 94.1	U3b2 52		U2b2 105.4

* Sampling or analytical problems.

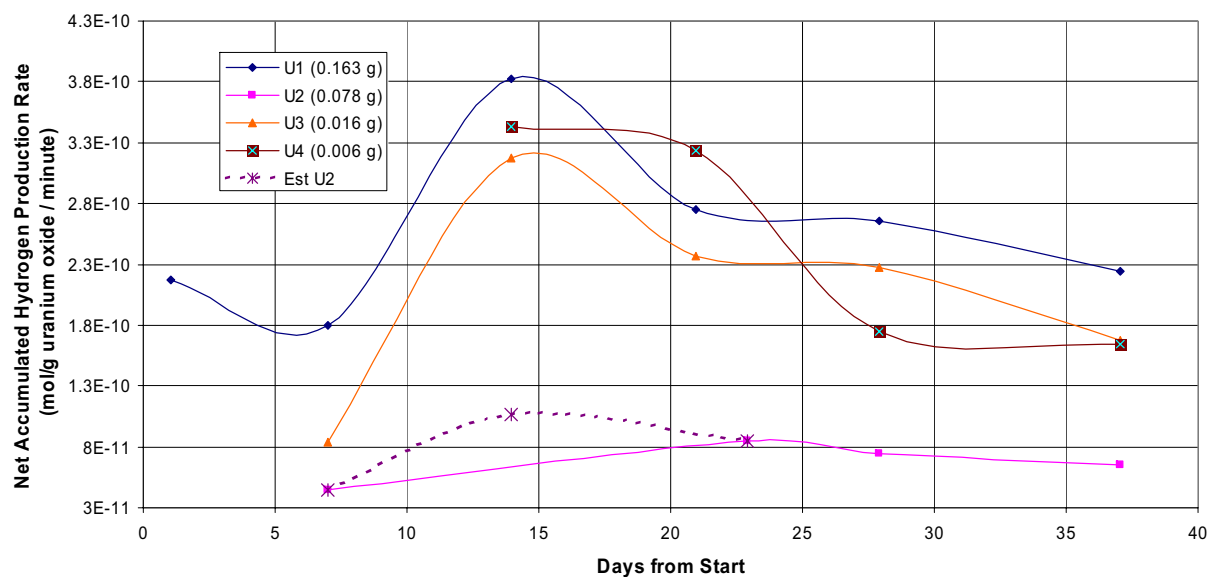


Figure G-4. Rates of net hydrogen production.

G-5. RESULTS AND CONCLUSIONS

The mass-normalized and volume-adjusted hydrogen rates are shown in Table G-5. The hydrogen concentrations were adjusted for the volumes purged from the system during sampling. These adjusted values were divided by the mass of uranium oxide embedded in the WAXFIX. In the first group of values presented in the table, rates accumulated at each sample point were averaged. The second group is the average of the rates for each generator from sample to sample. The latter rates vary more than the accumulated rates (the U2 generator excepted). The standard deviation value as a percentage of rates for the differential average is about twice that for the accumulated standard deviation values. For U2, the value is nearly the same. As will be discussed later, U1, U3, and U4 generators are consistent, and the values are meaningful. No hydrogen was detected in any of the samples drawn from the blank generator systems (the two that contained no uranium).

Table G-5. Mass-normalized and volume-adjusted net hydrogen generation rates.

Generator	Mass UO ₂ -g	Accumulated Average H ₂ Generation Rate – Gmol/(g. UO ₂)(min)	Standard Deviation	Differential Average H ₂ Generation Rate – Gmol/(g. UO ₂)(min)	Standard Deviation
U1	0.163	2.57E-10	7.04E-11	1.77E-10	1.06E-10
U2	0.078	6.72E-11	1.74E-11	3.96E-10	1.31E-11
U3	0.016	2.07E-10	8.71E-11	1.55E-10	1.18E-10
U4	0.006	2.51E-10	9.46E-11	2.00E-10	1.34E-10
B1	0	0	N/A	0	N/A
B2	0	0	N/A	0	N/A

The differences between the accumulated and differential averages are a result of computing the values in a different order using subtractions and time averaging. If the rates were constant over time, the values would have been more consistent for each method in both the rate and in the standard deviation. As shown below in Figure G-4, the rates of net hydrogen production were not constant over time. For the U2 generator, the indicated rates did not vary as much those of the other generators.

The upward trend in the rates shown in the graph is increasing from zero to high enough concentrations that can be more accurately analyzed by the gas chromatograph. Rates seem to peak at about Day 15, then decrease. This decrease could be a result of back reactions of the hydrogen with the WAXFIX or with other gases or materials in the generator system. It appears that the effect is common among the generators and the possible effects of sampling or analysis errors cannot be ignored. Back reactions consuming hydrogen should be hydrogen concentration-related and a lesser effect should be evident for lower mass generators than for U1, yet U3 tracks very well with U1 at about 1/10 the U2 mass and with less than 1/10 the hydrogen concentration.

Other possibilities for the downward trend could be preferential diffusion of the hydrogen from the generator systems or a slight loss of sensitivity of the gas chromatograph column to hydrogen caused by the presence of a contaminant that interferes with analysis, perhaps produced by the radiolysis.

The gas chromatograph instrument was checked against a quality assurance source before each set of measurements. On two occasions, no values for hydrogen were detected in any of the samples, and the generators were resampled. It is suspected that the samples were just air or that the gas chromatograph had a leak. On two other intervals, the sample from U2 produced no hydrogen and no argon, suggesting

lack of a true sample. The U2 sample problem was isolated as a bad adapter that was used to connect the sample syringe.

The values, with the exception of those from the U2 generator, are within one standard deviation (1σ) of each other on the average and thus are indicating essentially the same hydrogen generation rate per time per mass. Possible reasons for the differences shown by U2 are believed to be related to a gross difference in the particle size distribution, to an inefficient spreading of the uranium oxide particles in the wax, or to a mass weighing error.

With the particle size distribution, net G hydrogen values^a could be calculated from the data. A particle-size analysis on the uranium oxide was planned, but the instrument was not available.

The G value correlates the energy absorbed with the production of radiolysis product. In the present case, the product is hydrogen gas. The definition is 1 mol/100 MeV absorbed would be a G value of one. Figure G-5 correlates the expected G hydrogen with the particle size for alpha particles of the energy of the sample uranium oxide. To produce the graph, the linear energy transfer within the uranium oxide of average alpha particle energy had to be determined. This was estimated using the Bragg-Kleeman rule in Equations (G-1) and (G-2) (Vinson 2002):

$$R = R_{\text{air}} \left(\rho_{\text{air}} / \rho_x \right) \left(MW_x / MW_{\text{air}} \right)^{0.5} \quad (\text{G-1})$$

where:

- R = range in millimeter
- ρ = density
- MW = molecular weight
- _{air} = subscript for air
- _x = subscript for compound of interest.

$$R_{\text{air}} = 1.24 E_p^{-2.62} \quad (\text{G-2})$$

where:

- E_p = energy of the particle in megaelectron volts.

A range for the average energy alpha particle, 4.913 MeV, in air for the uranium used was calculated to be 34.72 cm (13.7 in.) using Equation (G-2). The linear energy transfer was then calculated from the range and the full energy of the average alpha particle using Equation (G-1). This value was determined to be 410.2 MeV/mm. Using the linear energy transfer, the hydrogen generation spreadsheet program previously mentioned and the average hydrogen generation data were developed. Figure G-5 shows the expected mass mean particle diameters, assuming a range of possible G values.

a. G hydrogen is the absolute number of hydrogen molecules produced per 100 MeV in a reaction.

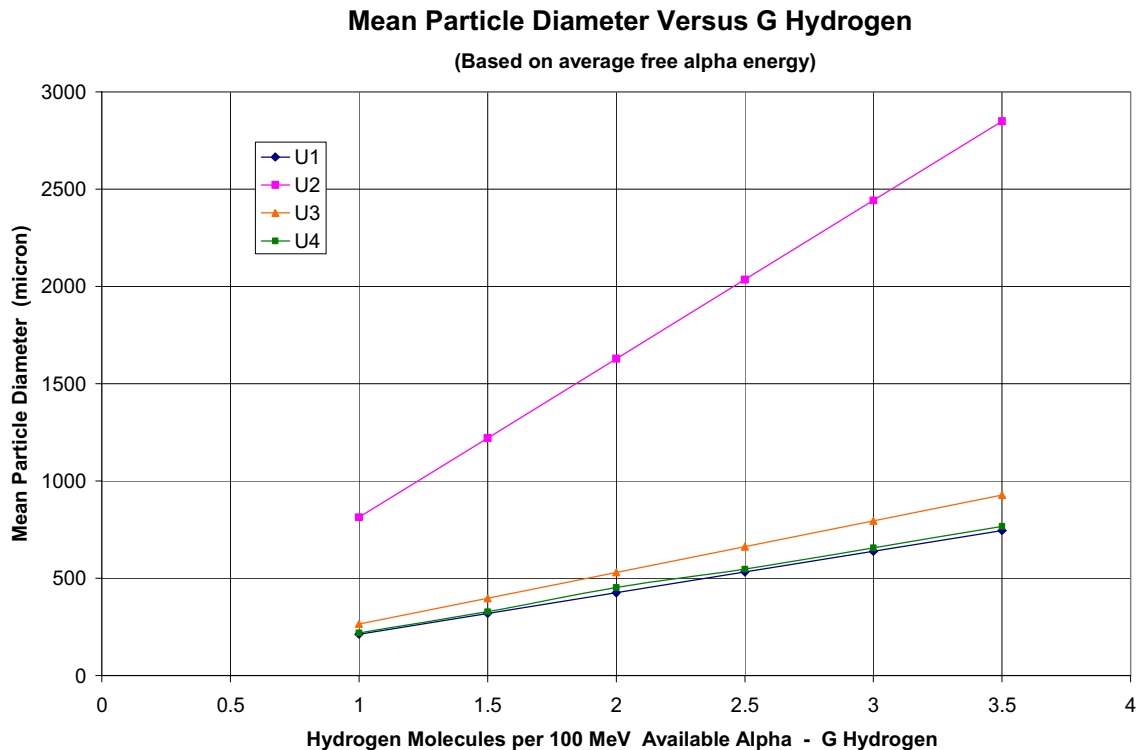


Figure G-5. This figure shows the expected mass mean particle diameter as a function of G hydrogen based on the measured and calculated data for each generator.

Examining Figure G-5, it is apparent that larger mass mean particle diameters would be required to fit the higher G values using the calculated linear energy transfer and the existing data. This may appear as counter intuitive, but note that the net hydrogen production rates and the linear energy transfer are fixed. At the indicated particle sizes, less than 4% of the alpha energy escapes the uranium oxide particles. To accommodate the total alpha energy, the particle size has to increase to absorb the energy. A much higher alpha flux would have been expected with a smaller mass mean particle diameter.

The plots from Figure G-5 show very reasonable agreement with the calculated fixed parameters and the hydrogen production rates for U1, U3, and U4. That is, the net G hydrogen values for reasonable particle size do appear to be within the expected range. The upper limit for the net G hydrogen is about 3.0 and the lower limit about 1.0.

The U2 is clearly an outlier. Not enough hydrogen was detected from generator U2 for the data to fit comfortably within the calculated parameters and the maximum physical particle size, unless the average mass mean particle size is over 1,000 microns and the G hydrogen is around 1.0. If the G hydrogen were to be higher than about 2.0, the mass mean particle size would have to be as high as the observed maximum for Figure G-3 (i.e., about 1,500 microns). There are problems with this generator that can be attributed to mass weighing error, gas sample dilution, uranium oxide contamination, or poor mixing with the WAXFIX.

From Figures G-2 and G-3, it is evident that the degree of mixing and particle size distribution of U1 is less favorable to hydrogen production than is that of the U1 generator.

Maximum particle diameters interpreted from the pictures, shown in Figures G-3 and G-4, are 1,000 and 1,500 microns, with an interpreted average for the clearly visible particles of 597 and 747 microns for U1 and U2, respectively. Note that the color of the wax of U1 has a larger dark area and, in general, is darker than U2. It was evident when the generators were prepared that U1 had a larger proportion of fines than U2, as is indicated in the photographs by the differences in the darker areas.

G hydrogen values for pure organics in the literature vary from 1.0 to over 6.0 (Bolt and Carroll 1963), but literature values sometimes do not consider recombination and are reported as gross values. For alkanes, numbers for gases are higher than liquids by a factor of about two. Recombination numbers, which account for the reverse reaction of hydrogen to water, show negative G hydrogen values from 1.0 to 1.7, depending on alpha energy density (Vinson 2002).

The highest average energy absorption density calculated for this experiment is $3.4\text{E}+06 \text{ MeV/cm}^3/\text{second}$. This is more than a factor of 10,000 less than values used in *Radiological Health Handbook* (Bureau of Radiological Health 1970). Because the heavy uranium oxide particles would not disperse properly in the wax, localized energy absorption densities occurred. Assuming that a net G hydrogen value of 2.0 is used, the gross value of G hydrogen could be 3.7. Using the average interpreted particle diameter for the picture for U1 as the mass mean particle diameter and the spreadsheet program, a net value for G hydrogen of 2.8 was calculated.

A special sample was drawn from U1 and U2 for higher organic gases (methane to propane were not detectable). Alkanes and some complex organics were detected, but the concentrations were below 1.0 ppm(v).

The test data are within the expected range and are a good estimate for the WAXFIX alpha radiolysis. Better and more complete test results can be obtained with controlled particle size, a better sampling, longer time intervals, and a more complete analysis of the gas.

G-6. REFERENCES

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